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Vitrification of Simulated LILW Using Induction Cold Crucible Melter Technology

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ABSTRACT

Vitrification destroys hazardous organics, and immobilizes heavy metals and radioactive elements to form a chemically durable and highly leach-resistant vitrified form. The vitrification process provides exceptional volume reduction and is attractive for minimizing disposal volume. A pilot plant test using an induction Cold Crucible Melter(CCM) fitted with an off-gas treatment system(OGTS) has been conducted to vitrify a simulated low-and intermediate-level radioactive waste(LILW) generated from Korean nuclear power plants. The CCM process is based on the use of a water-cooled metallic structure assembled in sectors which is transparent to the electromagnetic field supplied by a high-frequency generator. A solidified glass layer because of the water-cooled structure of the CCM protects the structure against corrosion. By creating the solidified glass autocrucible on the inner surface of the wall, corrosion damage to the steel in contact with the molten glass is prevented. In order to start-up the CCM, the glass frits were loaded in the CCM. The glass melting was initiated by heating of a short-circuited titanium ring in an electromagnetic field followed by ring burnout and incorporation of the titania in the glass frits. The melter has one drain that exits through the bottom. It is a direct bottom drain from the floor of the melt tank. It is sealed by the solidified glass layer and can be activated by removing the water cooling system. This drain is used if it is desired to drain the melter. The melter employs oxygen bubbling to promote mixing and to increase the melting rate. The bubblers are desired to produce a curtain of bubbles rising from the melter floor. In addition to mixing, the bubbling of oxygen tends to keep the melt well oxidized. The top of the melter is equipped with a number of ports. These provide access for feed, viewing, off-gas discharge, etc. The normal method of feeding is dry feeding through a feed pipe mounted through the top of the melter. The HFG power and operating frequency were applied in the ranges of 100~200 kW and 250~270 kHz, respectively. The simulated mixed waste vitrification test using the pilot scale plant consisting of the CCM and the OGTS at NETEC has demonstrated its good workability, reliability, and high productivity. The mixed waste was easily vitrified at a maximum rate of 20 kg per hour. The product quality of the glass such as chemical durability, phase stability, etc. was satisfactory. All regulated gases in the stack were well below the environmental regulation limits.

INTRODUCTION

Nuclear Environment Technology Institute (NETEC) has investigated and evaluated various efficient thermal treatment technologies for the low-and intermediate-level radioactive waste (LILW) generated from nuclear power plants (NPPs). NETEC has especially focused on a treatment technology for the LILW to have a large volume reduction effect which abates the rapid increase in saturation of on-site storage capacity and disposal costs, enhances the stability of the waste form, and can treat all waste streams generated from Korean NPPs.

In the early 1994, an application of vitrification technology to the treatment of the LILW was concluded as the most promising technology because it can achieve large waste volume reduction, create a durable waste form and destroy organic compounds more effectively than other competing technologies such as cementation, polymerization, and ceramic formation. The vitrified radioactive waste is expected to remain stable in the repository environment more than one million years because this technology is to chemically incorporate the radionuclides in the LILW into the durable glass matrix, but not an encapsulation technology. Therefore, the safety of the repository can be greatly enhanced. In addition, vitrification technology contributes to remarkable waste volume reduction factor from 33 to 176 for the simulated LILW compared to the initial bulk volume of the waste and results in efficient, and prolonged utilization of the disposal site in Korea.

The multi-step R&D projects to vitrify the Korean LILW have been performed. A feasibility study of the application of vitrification technology to the LILW has been conducted by aiming laboratory tests to assess the melter technologies, to examine how innovative high temperature technologies could be implemented to achieve a large volume reduction, and to evaluate and compare these technologies from a technical and economic viewpoint from October, 1994 to October, 1995. As a result of the feasibility study, the cold crucible melter(CCM) with an off-gas treatment system(OGTS) was selected for vitrifying the combustible radioactive waste. And its technical and economical aspects were assessed to be superior to current technologies[1].

An international joint research and development program started in July, 1996 with the support of SGN(AREVA group, France) and MOBIS(HYUNDAI group, Korea) to develop the cold crucible vitrification of the Korean LILW using a pilot scale CCM through a multi-step program. The first step of the program, completed in 1998, was dedicated to orientation tests to optimize the processing of the Korean LILW in the CCM and to design the off-gas treatment system. The second step of the joint collaboration, completed in September, 1999, was devoted to the design and construction of the pilot scale vitrification facility in Daejeon, Korea. This pilot scale vitrification facility as shown in Fig. 1 has been in operation since October, 1999. And the pilot test has been performed by NETEC with supports of SGN and MOBIS. Finally, the pilot scale vitrification tests have been intended to finalize the design parameters, design options, license data, and so on. As a part of the project, the glass formulation study has been conducted in parallel with the pilot scale vitrification tests in the laboratory and the pilot facility using simulated LILW by considering the processability and the durability of the waste glasses, and volume reduction effect.

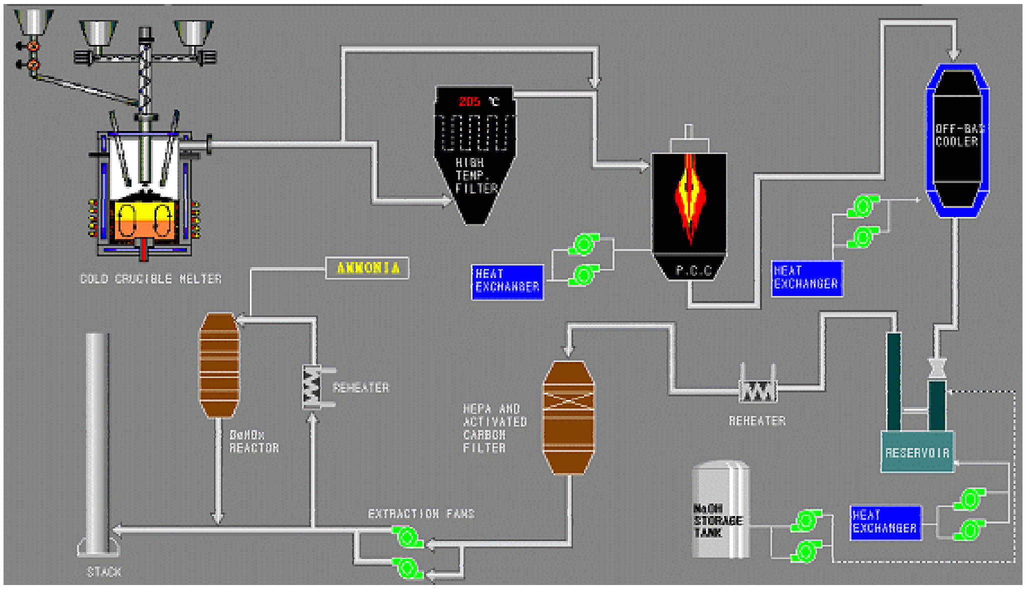

Fig. 1. The pilot scale vitrification facility consisting of induction cold crucible melter and OGTS

DEVELOPMENT OF INDUCTION COLD CRUCIBLE MELTER SYSTEM

In the cold crucible concept, the crucible itself is a simple, robust structure made of plain stainless steel sectors, cooled by water as shown in Fig. 2. Electrical currents are directly induced into the glass melt from an external high frequency inductor. The molten glass is directly heated while the crucible wall is cooled. This cold structure causes a thin layer of solidified glass material to coat the surface of the crucible in contact with glass, thereby protecting it from the corrosive melt. Since the energy source is outside the melter, the recurring problem of electrode corrosion is avoided. In addition to providing a considerable lifetime for the melter, this technology overcomes the temperature limits imposed in more conventional systems with Inconel electrodes(1,050□1,150□). This technology accommodates corrosive glasses or a refractory melt(more than 1,150□) that could not otherwise be processed. The glass is poured periodically through a cooled bottom drain valve.

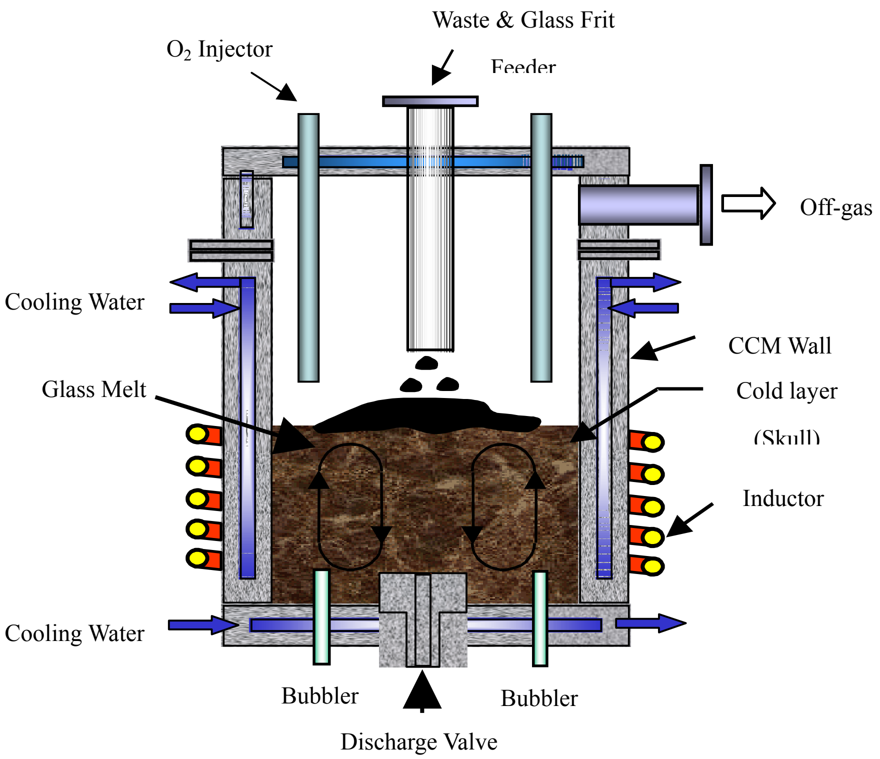

Fig. 2. Cross sectional view of the CCM developed by NETEC

The structure, in sectors, helps to develop an electromagnetic field in the crucible. The melter employs oxygen bubbling to promote mixing and to increase the melting rate. The bubblers are desired to produce a curtain of bubbles rising from the melter floor. In addition to mixing, the bubbling of oxygen tends to keep the melt well oxidized as shown in Fig. 3. The top of the melter is equipped with a number of ports. These provide access for feed, viewing, off-gas discharge, etc. The dry and wet feeding methods are available through a feeder mounted through the top of the melter.

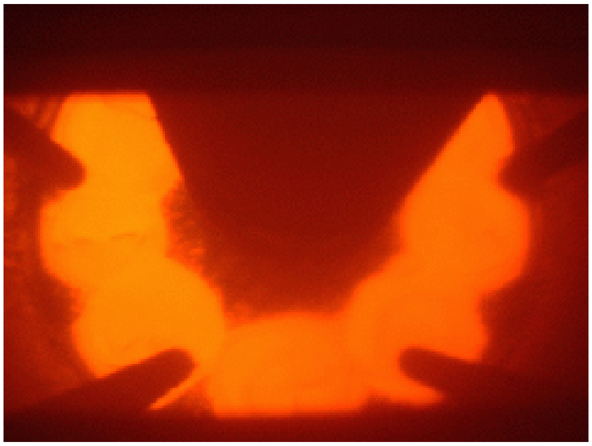

Fig. 3. Photograph of oxygen bubbling using 9 bubblers installed at bottom of CCM

The off-gas treatment system(OGTS) for the pilot scale vitrification facility consists of a pipe cooler, high temperature filter(HTF), post combustion chamber (PCC), packed bed scrubber, air reheater, HEPA filter, and SCR units as shown in Fig. 1. This off-gas system was designed to treat particulate, aerosol, and acidic gaseous emissions. The OGTS was operator assisted, controlled by ladder logistic alarm levels while the critical parameters (temperature, pressure and current/voltage) were continuously monitored by sensors directly interfaced to a PC. Off-gas monitoring was performed in two distinct modes: Continuous Emission Monitoring(CEM) for the gaseous compounds O_2, CO, CO_2, NO_x, SO_x, and HCl and standard isokinetic sampling for dust. During the pilot scale vitrification test, concentration of O_2, CO, NO_x, SO_x and C_xH_y were directly analyzed at the outlet of HTF by portable gas analyzer(PGA). Sampling of dust was conducted at the outlet of the CCM and analyzed the concentration of dust.

PILOT SCALE VITRIFICATION TESTS

A lot of pilot scale vitrification tests were carried out with simulated dry active waste(DAW), ion exchange materials(IEX) including high and low activity resin(HA/LA resin), and zeolite, and mixture of the DAW and IEX, for the major waste streams generated from Korean nuclear power plants. The result of the pilot tests proved that environmentally stable vitrified form for all the wastes could be produced. All regulated gases in the stack were well below the environmental regulation limits. And, a remarkable waste volume reduction effect was achieved. The brief results of the pilot scale tests for the major waste streams are as follows.

Operability

In order to start-up the CCM, about 70kg of glass frit was loaded in the CCM. The glass melting was initiated by heating of a short-circuited titanium ring with 38cm in diameter in an electromagnetic field followed by ring burnout. Then the titania was incorporated into the glass frit and simultaneously generated the heat energy based on the following equation, $Ti + O_2 \rightarrow TiO_2 + 890kJ/mol$. In the case of using about 80g of titanium ring and ignition for 10 second, about 150kW was supplied to the environment of the CCM. After the start-up completed, the HFG power and operating frequency were applied in the ranges of 100□150kW and 250□270kHz, respectively. The optimum feeding rates of the wastes and the proper amount of excess oxygen were determined through the pilot tests. The control of the glass melt at 1,150±30□ was found to be easy during the waste feeding. The waste accumulation on the glass melt surface was not observed while the waste and glass frit were simultaneously fed into the CCM. Three more than hundred hours(two 120 and one 180hrs) continuous tests were successfully performed for vitrifying mixture of the DAW, HA resin, LA resin and Zeolite simulated wastes being generated from Ulchin 5&6 units. The pilot scale vitrification facility has showed their integrity. The glass melts were easily drained through a direct bottom drain valve located on the floor of the CCM.

Physical Properties

The vitrified forms produced from the tests were homogeneous without secondary phase formations such as sulfate, sulfide, metal, etc. according to SEM/EDS analysis. The compressive strengths of two heat treated vitrified forms for mixture of the DAW and HA resin were in the ranges of 3,208□3,316kg/cm^2 (47,159□48,747 psi). These values were up to 90 times higher than the 500 psi required for the cementation by the US NRC[2].

Chemical Properties

According to 7-day PCT using three vitrified forms sampled from initial, middle, and final stages of the test for the mixture of DAW, LA resin and Zeolite, the chemical durability was satisfactory. Especially, Fig. 4 shows the 7-day PCT result for vitrified forms for the W1 waste. The leach rates of 7-day PCT for Si, B, Na, and Li in three samples were below 0.5g/m^2. The results showed that the vitrified form had a lower leachability than the benchmark glass(SRL-EA) used in assessment of radioactive waste vitrification in the USA[3].

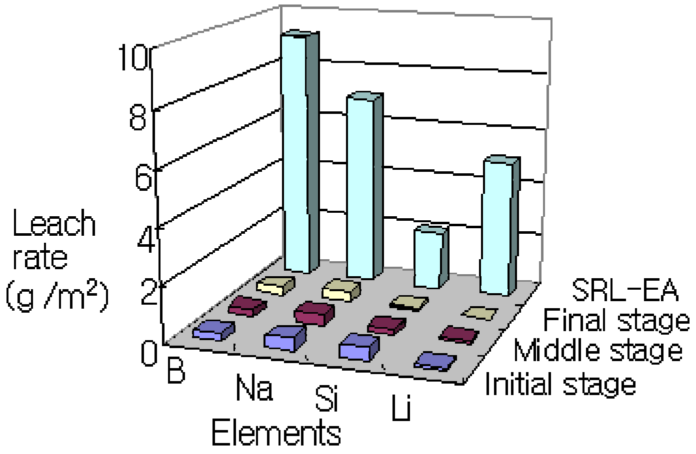

Fig. 4. 7-day PCT results for initial, middle, and final stage vitrified forms produced from vitrification process for the W1 waste

Off-gas Properties

The hazardous off-gases(CO, NO_x, SO_x) generated during the waste feeding onto the glass melt were completely treated in the OGTS and their concentrations at stack were well below the environmental regulation limits. Also, the concentration of dioxin at stack was about 1/278 of the environmental permissible emission limit.

Volume Reduction Effect

The volume reduction factors evaluated based on the initial bulk volume of the waste fed and vitrified form produced were 176, 33, 74, and 84 for the DAW, W2 waste(DAW+HA Resin+LA Resin+Zeolite), mixture of the DAW and HA resin, and W1 waste(DAW+LA Resin+Zeolite), respectively.

CONCLUSIONS

A successful R&D on vitrification of NPPs wastes has been made through the pilot tests using the induction cold crucible melter developed by NETEC. In order to start-up the CCM system, the glass frit was loaded in the CCM. And the glass frit melting was initiated by heating of a short-circuited titanium ring in an electromagnetic field followed by ring burnout and incorporation of the titania in the glass frit. The melter employs oxygen bubbling to promote mixing and to increase the melting rate. The bubblers are desired to produce a curtain of bubbles rising from the melter floor. The vitrification tests for the simulated wastes using the CCM and the OGTS have demonstrated the good workability, reliability and high productivity, and has simulated the behavior of a commercial vitrification facility as closely as possible. The simulated wastes were easily vitrified in the range of 15□25kg per hour. The product quality of the glass such as chemical durability, phase stability, etc. was satisfactory. All regulated gases in the stack were well below the environmental regulation limits. Through these tests, the remarkable waste volume reduction factor of greater than 33 compared to the initial bulk volume of the waste could be gained. Accordingly, it is expected that the vitrification technology will not only enhance the safety of the waste disposal repository, but also greatly contribute to the further promotion of Korea's nuclear power generation program.

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