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IN JAPAN

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ABSTRACT

Liquid-fed Joule-heated ceramic melter(LFCM) process for the vitrification of high-level liquid waste(HLLW) is now under development by Power Reactor and Nuclear Fuel Development Corporation(PNC) in Japan. All developmental works are focussed on the vitrification plant which is in the stage of design improvement in succession to the detailed design finished in 1984. The construction of the plant will be started in late 1987. Major development items in process technology in recent years are improvements of both the design of a ceramic melter and performance of melter off-gas cleanup system. A new engineering-scale ceramic melter with a 915 MHz microwave heating device has been in operation since September 1985. Melter dismantling technique and numerical flow analysis code and others are also being developed as the part of melter technology. In the off-gas technology, air-film cooler at the outlet of the melter off-gas, high efficiency mist eliminator and wet type electro precipitator were developed and checked the performance. In the Chemical Processing Facility(CPF), hot vitrification and product characterization works are continued.

INTRODUCTION

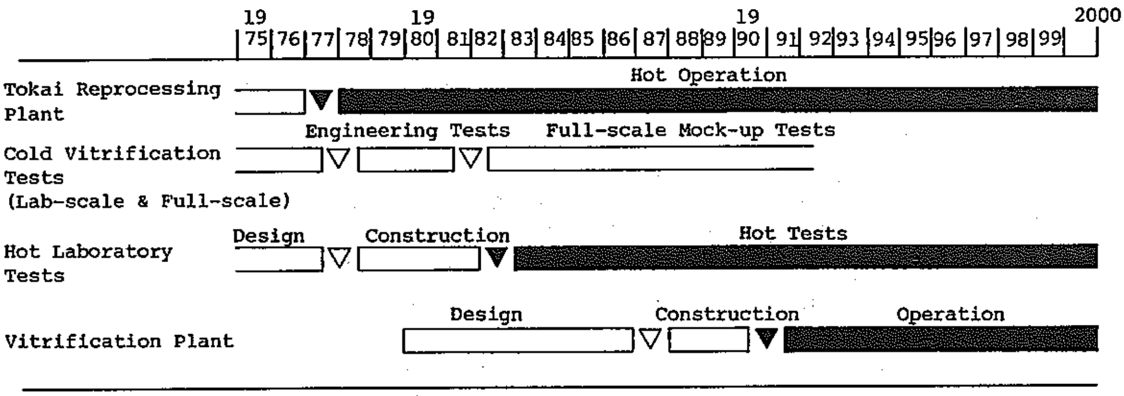
Development of vitrification technology in PNC was started in 1976. Many activities on LFCM process technology have been carried out in the operation of Engineering Test Facility(ETF) and Mock-up Test Facility(MTF) since 1980 and 1982, respectively, and have provided design basis for the process design. In partivular, full-scale cold mock-up vitrification tests have been performed in MTF.[1] Laboratory-scale hot vitrification tests were started in December 1982 in CPF, and nine runs have been already made untill September 1986 along with the characterization of the waste glass.

Design of the vitrification plant has been successively carried out since 1980. The detailed design of the plant was completed in 1984. At present, the design improvement is being made for the reduction of the construction cost and for the licensing which is going to be applied in this year. The construction of the plant will be started in late 1987. Table 1 shows a schedule on the development of HLLW vitrification technology in PNC.

In this paper, outline of the vitrification plant and major development items in process technology in recent years are described.

Table 1

Schedule for the development of HLLW vitrification technology[2]



OUTLINE OF THE VITRIFICATION PLANT

Vitrification process

Figuer 1 shows a flow of the vitrification process.[2] The HLLW is transfered from the Tokai Reprocessing Plant to a receiving tank. Elemental and radioactive analysis are carried out for process and product quality control. The HLLW is pretreated to adjust the composition by the addition of chemicals and/or by concentration in an evaporator when required. After the pretreatment, HLLW is fed into a melter continuously using a two-stage airlift. Glass fiber cylinders are used as glass additive for melting. The HLLW is soaked into the cylinder just before it is fed into the melter. The molten glass is discharged periodically through a metallic nozzle located at the bottom of the melter into a canister. During the discharge, the weight of the glass in the canister is successively measured by load cells. The filled canister is subsequently cooled, transfered to the welding position, and a lid is welded by a TIG welder to seal the canister. After being decontaminated by high-pressure water jet spray and wire brushing and being inspected, packages are stored in forced-air cooling storage pits.

Melter off-gas is cleaned by dust scrubber, venturi scrubber, perforated plate water scrubber, high efficiency mist eliminator, ruthenium adsorber (silica gel), HEPA filter and so on. The off-gas from the receiving tank, the evaporator of HLLW and the secondary liquid waste treatment system is cleaned in a separate system.

Specification and features of the plant

Major specification of the vitrification plant is shown in Table 2. The capacity is equivalent to the reprocessing of 0.7 ton of heavy metals per day. This plant employs fully remote operation and maintenance in a large vitrification cell with low flow ventilation. All equipment in the cell are placed in standardized rack-mounted modules. As remote maintenance tools, in-cell cranes and two-armed servo-manipulators are equipped in the cell, and ITV is the main viewing system.

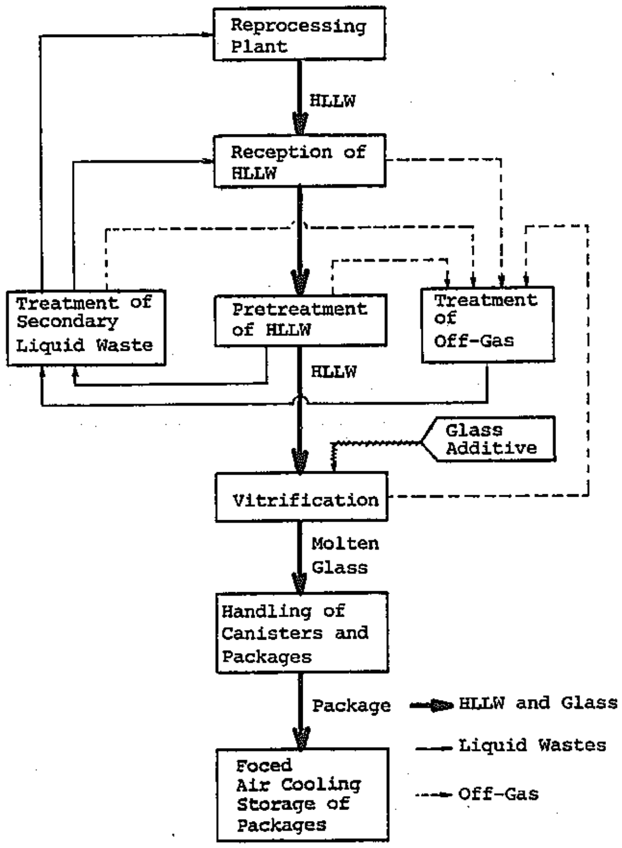


Fig.1 Block flow diagram of the vitrification process[2]

MAJOR DEVELOPMENT ACTIVITIES

Major process technologies which have been developed are listed in Table 3.

Glass composition

Glass composition has been studied and improved from the beginning of the development. At the earlier stage, glass composition with about 43 % of SiO_2 was selected as a reference. Recently, glass with higher SiO_2 content is optimized from the results of characterization of the glass.

TABLE 2

Specification of the plant

HLLW	Burnup of spent fuel	28,000 MWD/MTU
	Specific power	35 MW/MTU
	Initial U-235 content	4 wt%
	Cooling time before reprocessing	0.5 Y
	Cooling time before vitrification	5.5 Y
	Volume of HLLW generated (design base)	0.5 m^3/MTU
Glass composition (standard)	Glass additives	75 wt%
	Waste oxides	25
	FP	10
	Na O	10
	Others	5
Package	Canister	SUS 304L
	Glass	300 Kg
	Heat generation rate	1.4 KW
	Radioactivity	4 X 10^5Ci
Capacity	Glass production rate	about 9 Kg/hr
	Package production rate	140 packages/y
Storage	Forced-air cooling	

Preteatment of HLLW

Denitration of HLLW with formic acid was studied in PNC for several years to suppress the volatilization of ruthenium during melting. However, since the operation of feed system for the denitrated HLLW is difficult owing to deposits, and volatiled Ru will be removed in the off-gas system, it is decided the denitration should be discarded in the vitrification plant. The control system for the constant feed rate of HLLW under air-pulsation conditions is now being tested using the two-stage airlift technique.

Melter technology

Melter bottom structure

Full-scale tests have been performed through the operations of test melters to establish the melter bottom structure. In order to avoid the operational problems caused by accumulation of electro-conductive sludge, it is concluded that melter bottom refractory should be sloped and equipped with a bottom drain nozzle. The slope of 30 to 60 degrees is desirable to facilitate the discharge of the deposits without any operational difficulties.

Table 3

Major development items related to vitrification process

Subjects	Development items
Glass composition	Glass composition with higher SiO_2 content
Pretreatment	Control system for the constant feed rate of HLLW by using two-stage airlift
Melter	Melter bottom structure Bottom freeze valve with induction heating Glass level detection Continuous feed system for glass fiber additive Dismantling technique of the melter after its operation life time Extention of the service life time of the melter Numerical flow analysis code of the melter Microwave heating device
Off-gas treatment	Prevention of melter off-gas pipe plugging Improvement of cecontamination factor for submicron particle
Canister	Lid welding by non-filler TIG technique Decontamination and inspection technique

Glass additive

In the development of glass forming materials, powdered and granular glass (beads) additives have ever been attempted to the LFCM process, and feed systems of these glass additives have been completed.

For the stable operation of melter, however, it was found that the reduction of the particulate entrainment in the off-gas stream from the melter is important to prevent the plugging of the off-gas pipe. Cylindrical glass fiber additive has been developed in place of the former two types for this purpose, and in recent operation of the melter, glass fiber additive is used as the feed material in the LFCM process.
Specification of glass fiber additive is shown in Table 4. Figure 2 shows the particle entrainment rate in the off-gas stream. The entrainment of particle in the off-gas is reduced up to 1/10 compared with the feed of beads.

Table 4

Typical specification of glass fiber additives

Property	Binder type	Sintered type
Dimension (typical), mm	ϕ70 X ℓ70	ϕ70 X ℓ 70
Weight per piece (typical), g	38	50
Bulk specific gravity	0.18	0.20
Fiber element, diameter, microns	10～15	8～12
Binder or Coating additives	SiO_2-CaO-$A\ell_2O$-Li_2O-Na_2O	H_3BO_3
Maximum retention volume, ml/g		
at 330m /min of feed	>2.5	>4
immersed	>4	>4

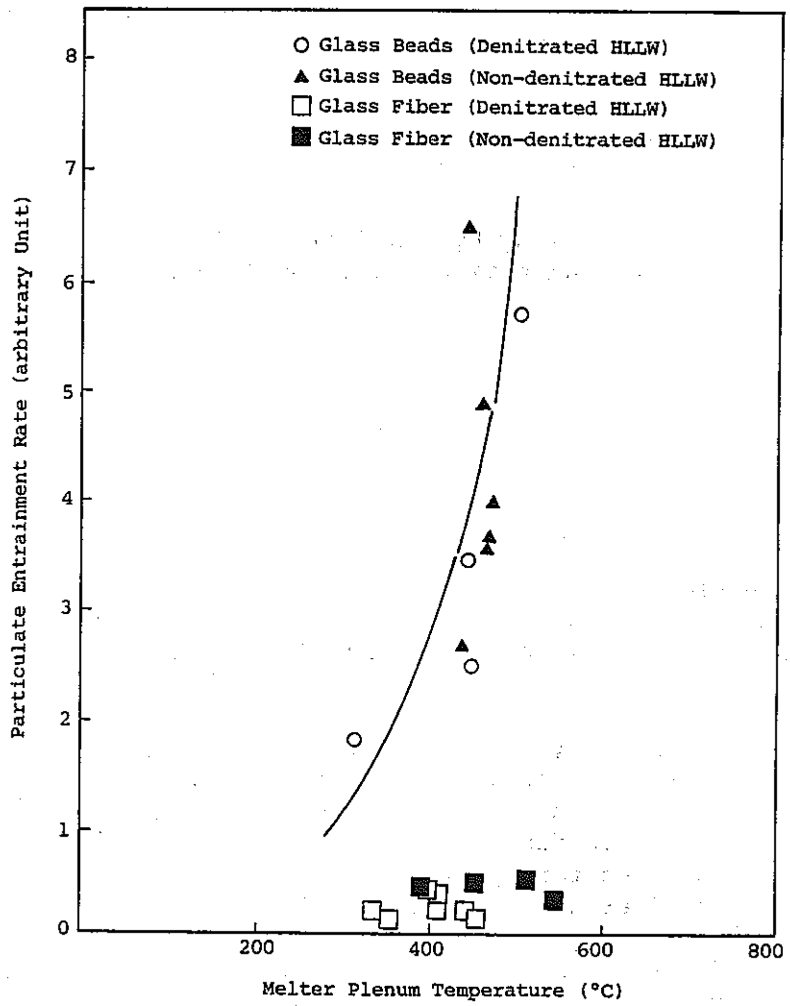


Fig.2 Comparison of particulate entrainment rate in off-gas from melter between glass beads and glass fiber additives

Glass drain

Freeze valve based on both direct current heating with mechanical shear system and stopper, and induction heating have been compared with consideration of formation of glass string at the termination of the glass drain. From the standpoint of termination of glass draining, two-stage induction heating is evaluated to be suitable because of the simplicity of the operation.

Level detection

Electrical resistance, direct contact and microwave level detection system have been tested. The former two types of leveler have been well estimated, and will be adopted in the melter of the vitrification plant. Further development is required for microwave leveler.

Heating up

Heating up technique with conventional SiC resistance heater is well developed. As a technique both to increase the processing capacity and to increase the maintenance ability, microwave heating is under development as a promising future alternative method. Photograph 1 is a new engineering-scale ceramic melter with microwave heating device (915 MHz). This was installed September 1985 and has been well operated without any difficulties.

Other related technology

As an attempt to extend the service life time of the melter, more corrosion resistance metallic and ceramic materials have been studied. A new ceramic material which has both adequate electrical resistance and three to four times more ocrrosion resistance than fused-cast K-3 was successfully found in a laboratory.

Melter dismantling technique, inspection device and numerical flow analysis code of ceramic melter are also under development.

Off-gas treatment

In the development of off-gas treatment techniques, an air-film cooler, high efficiency mist eliminator(HEME), and wet type electro precipitator(WEP), are being tested in recent years. Air-film cooler is based on almost the same concept as developed in SRL[3]. The test in MTF showed the good performance; there was little deposits on the inner surface of the melter off-gas pipe. A HEME showed the high decontamination factor of more than 100 in the melter off-gas treatment system. A WEP has been tested in MTF since October 1985 as one of advanced off-gas treatment equipment. It is found that DF for the particle with a diameter of 0.1 μm exceeds 1000 when adequate voltage is applied between electrodes. A submerged bed scrubber is also under development. Behavior of volatiled Ru and other elements are being studied in MTF and CPF.

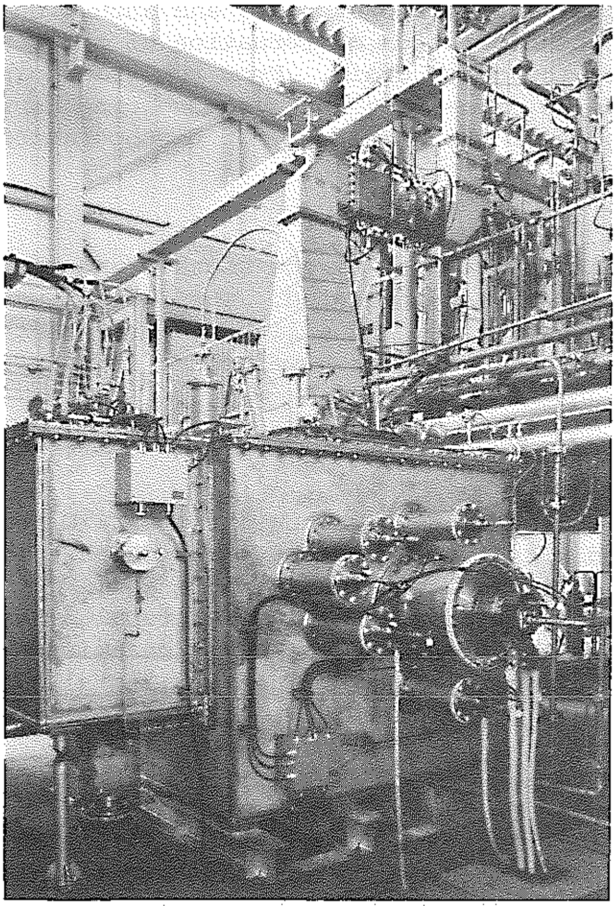


Photo. 1 New engineering-scale ceramic melter

Canister handling

The design of canister is completed. Temperature, deformation, and other characteristics at a glass filling have been measured. Drop impact tests for filled canister (package) were completed.

Lid welding is based on a TIG welding method without feeding filler wire. The quality is assured by controlling the welding parameters. Extensive welding tests are being performed to establish the parameters.

Hot decontamination tests are now conducted in CPF by using samples taken from the samll canisters filled with radioactive glass. Enough decontamination factor is attained by high-pressure water jet spray and wire brushing.

CONCLUSION

Many R&D activities have been carried out to support and improve the design, construction and operation of the vitrification plant. Design of the vitrification plant is now at its final stage, and it is planned to start the construction in 1987 and hot operation in 1991. Further R&D is continued for the demonstration and advancement of the vitrification technology in Japan.

REFERENCES

1. N. SASAKI, M. KARINO, H. OKAMOTO, H. KASHIHARA, and M. YAMAMOTO, "Solidification of The High-Level Liquid Waste from The Tokai Reprocessing Plant", Proc. Fuel Reprocessing and Waste Management, Jackson, Wyoming, August 26-29, 1984, Vol.1, p.1-147, American Nuclear Society (1984).

2. K. UEMATSU, "Design of the Vitrification Plant for the HLLW Generated from The Tokai Reprocessing Plant", Waste Management 86, Tucson, Arizona, March, 1986.

3. John L. Kessler, Chris T. Randall, "Performance of A Large Scale Melter and Off-Gas System Utilizing Simulated SRP DWPF WASTE", Proc. Symposium on WASTE MANAGEMENT, Tucson, Arizona, March 11-15, 1984, Vol.1, p.279-284, American Nuclear Society (1984).