WASTE TREATMENT EXPERIENCE AND FUTURE PLANS IN TOKAI REPROCESSING PLANT

A. Aoshima, K. Tanaka
Waste Management Division, Reprocessing Center in Tokai Works
Japan Nuclear Cycle Development Institute (JNC)
4-33 Muramatsu Naka-gun Tokai-mura Ibaraki, 319-1194 Japan

ABSTRACT

In the twenty seven years’ operation of Tokai Reprocessing Plant (TRP), liquid and solid wastes generated in the reprocessing process were treated adequately and stored safely in the on-site storage facilities. Among the waste treatment technologies which were used in TRP, JNC is making an effort to develop two solidification technologies of high radioactive liquid waste (HALW) and concentrated low active liquid waste (CLALW) respectively. For vitrification, JNC constructed the Tokai Vitrification Facility (TVF) in 1992 and has operated TVF for over ten years. During 2002 to 2004, JNC exchange the first glass melter for improved one which is changed the melter bottom design to have better drainability of glass. For solidification of CLALW, JNC constructed the Bituminization Demonstration Facility (BDF) and had bituminized CLALW. But, there was the world trend to prohibit radioactive wastes from sea disposal, JNC started the concept design of the new facility 1989 and started construction of the Liquid Waste Storage Facility (LWTF) in 2002. In LWTF, the ROBE solidification technology (Borsaure Einegung Anlage) which produces borated solids as interim products is used. The construction of LWTF will be finished in 2006 and enter operation.

INTRODUCTION

Japan Nuclear Cycle Development Institute (JNC) has operated the Tokai Reprocessing Plant (TRP) successfully for about thirty years since the start of its hot test operation in 1977 and the amount of reprocessed spent nuclear fuels mainly from the Japanese utilities reached over a thousand and sixty tones in total. TRP was designed to have the capacity of 0.7 tons U/day and treat the LWR spent fuels burned up 28,000 MWD/MTU in average.

In TRP, liquid and solid wastes generated in the reprocessing operation are classified into high and low radioactive wastes categories and treated by adequate method; high radioactive solid waste (HASW) and noncombustible low radioactive wastes (LASW) were stored in each storage facilities safely and combustible low active solid waste was incinerated. High radioactive liquid waste (HALW) was vitrified and low radioactive liquid waste (LALW) was evaporated and distillated flow was released to the sea after strict composition checking of satisfying the environmental release limits. Concentrated low radioactive liquid waste (CLALW) was bituminized in the Bituminization Demonstration Facility (BDF) until the fire and explosion incident of the BDF occurred in 1997. After the incident, the CLALW was stored in the tanks in the existing facilities and the new storage facility (LWSF:

Liquid Waste Storage Facility) built in 2003. Used waste solvent such as 30%TBP-dodecane was also solidified in the Solvent Treatment Facility (ST) by mixing with plastic.
All waste treatment operations were successfully and much operational knowledge and experience were accumulated.

In these operations, there are two important fields which JNC is trying to put stress on now: (1) technology improvement of vitrification and (2) construction of new facility for solidification of CLALW.

In TRP, HALW from the main process is vitrified in the Tokai Vitrification Facility (TVF). TVF was constructed and entered the hot operation in 1995 and produced 150 canisters until the end of 2004.

The type of TVF melter is the liquid fed joule-heated ceramic melter (LFCM) and the melter is designed to exchange every five years in the maximum operation mode because of the structural material corrosion.

On March of 2002, the damage of one main electrode occurred and JNC immediately accelerated the failed melter exchange activity to the new one. It was estimated that the electrode damage was caused by accumulation of the noble metals which are suspended in the glass-waste mixture. So, the design of the second melter was modified from the first one to have more effective discharge ability of the platinum group metal compounds. In parallel to the exchange activity, JNC has started detail study on the noble metals' behavior in the molten glass and the method for managing accumulation of the noble metals in the melter. JNC will advance this R&D activity and will apply gained knowledge

Fig. 1. Bird's view of the Tokai Reprocessing Center

to the future melter.

JNC has developed the melter dismantling technology which uses the YAG laser system for cutting structural materials and has finished the device design and fabrication. The first melter dismantling will be started in 2005 and it also includes detail investigation of the failed electrode.

JNC also has started new technology development for glass volume reduction.

The new facility (LWTF: Liquid Waste Treatment Facility) for solidification of concentrated low radioactive liquid waste is under construction in the site. In LWTF, CLALW is divided to major very low radioactive flow and minor middle level radioactive flow by coprecipitation / ultrafiltraton and ion exchange resin. Both wastes are solidified as intermediate solids by mixing with borate. The LWTF is also equipped with an incinerator for combustible solid wastes including plastics.

The construction of the LWTF started in 2002 and will finish in 2006.

VITRIFICATION

Facility Description

TVF located on the south side of the TRP site and the high radioactive liquid waste (HALW) transferred from the High Radioactive Liquid Waste Storage Facility (HAW) is vitrified and poured into a 110 L canister in TVF.

The TVF melter is designed to have capacity equivalent to the reprocessing capacity of 0.7 tons MU/day of TRP, which treats the LWR spent fuels burned up 28,000 MWD/MTU in average.

TVF has characteristic features in maintenance system designs: the fully remote maintenance system, which makes equipment exchange work so saving comparing to direct maintenance. In the fully remote maintenance system, all main equipment besides the melter is mounted in racks, which is installed in the large "the vitrification cell" and the all equipments are remotely maintained by using in-cell crane and the two-armed manipulators. The melter is also installed in the vitrification cell by bolting on the base framework.

TVF consists of two buildings and the stack. Two buildings are the process building and the administration building.

The process building has two floors underground and three above ground. In the underground part of the process building, there is the vitrification cell where almost main process equipment such as the receiving tanks, off-gas treatment equipment etc. are installed. Most equipment in the vitrification cell is mounted in the standardized racks with the dimensions of 3 m x 3 m x 6.5 m. TVF incorporates seven racks and the melter mounted on the support frame.

They are arranged along the both sides of the cell wall and the racks and the melter are remotely removable.

For maintenance, two overhead 20t cranes and two BSM (bilateral servo-manipulator) systems are disposed. These systems are operated fully and remotely from the control room. Images from the radiation hardened ITV cameras and the radiation hardened high definition TV camera (HDTV) mounted on the cranes and the BSMs are utilized by operators to maneuver the remote maintenance system. TVF has the storage cell for storing canisters containing solidified glass. The storage cell has the seventies pits which have the storage capacity of 420 canisters in total. Exit air from the TVF area ventilation is fed as cooling air to the pits and exhausted through the stack.

Process Description

Feeding the dissolved solution to the mixer settlers which are equipped in the separation process of TRP, the waste stream (raffinate) which contains almost radioactive nuclide besides uranium and plutonium flows into the subsequent evaporation process. After evaporation of the raffinate about ten times, the concentrated HALW is transferred to HAW and stored.

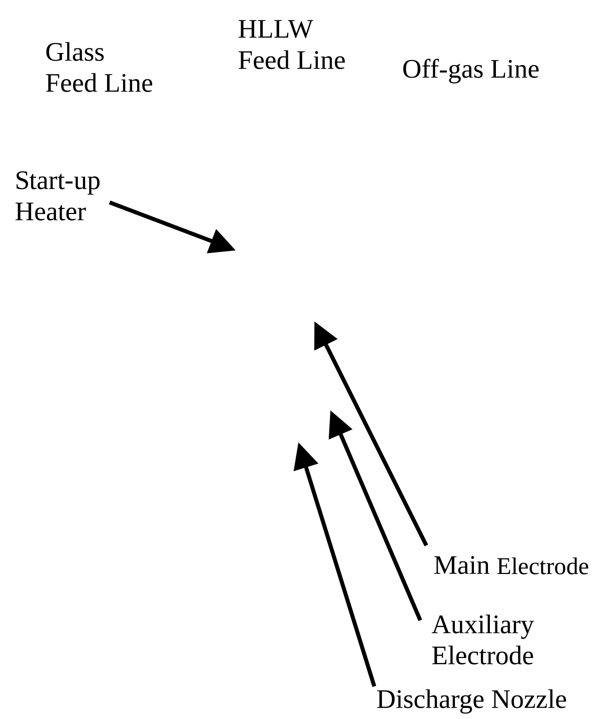
HALW is transferred from HAW to a receiving tank of TVF. Chemical compositions and radioactivity are analyzed for process and product quality control. HALW is pretreated to adjust the composition by adding chemicals like sodium and concentrating by an evaporator.

After the pretreatment, HALW is transferred to the melter continuously using two airlifts. (Fig.2)

Glass fiber cylindrical cartridge is used as a glass additive for melting. HALW is soaked into a cartridge before it is fed into the melter. The glass melted at the temperature of 1150 . is poured into a canister periodically through a discharge nozzle located at the bottom of the melter. The discharge nozzle is heated and cooled by switching the induction coils. The bottom part of the melter was designed to have the 45°sloped bottom to get good draining behavior of the glass. During the pouring, the weight and glass level are successively measured by load cells and a gamma-ray device, respectively.

The filled canister is subsequently cooled before transfer to the welding position and the lid is welded by a TIG welder remotely to seal the canister. After being decontaminated by high pressurized water jet spray and wire brushing, inspection of contamination and containment are carried out. Finally, canister is stored in the storage pits.

Off-gas from the melter is cleaned by a submerged bed



scrubber (SBS), a venturi scrubber, a perforated plate type water scrubber, a high efficiency mist eliminator (HEME), and subsequent filter process such as a ruthenium absorber, an iodine absorber and HEPA filters. The off-gas is mixed with area off-gas and finally exhausted to the atmosphere through the stack.

Operation Experience

The construction of TVF started in mid 1988 and finished in 1992. Followed the cold test, TVF started the hot test operation in 1994. In the beginning of the hot test campaign accumulation of the draining glass in the coupled device between the melter and the canister was occurred during the third glass draining to the canister. It was cleared that the accumulation was occurred by inadequate glass temperature control of the bottom part of the melter. So, changing the temperature control value to the adequate one, the test operation was restarted and produced 22 canisters successfully.

Through the hot test operation, the special operation method, "the low temperature operation mode" which keeps temperature of the melter bottom glass around 800-900 . was evaluated. The special operation mode adjusts the glass temperature to avoid accumulation of noble metals to the melter bottom by increasing the viscosity of the glass.

Though successful operation continued, the fire and explosion incident in the bituminization demonstration facility of TRP in 1997 interrupted the TVF operation till 2000. On June of 2000, TVF restarted and continued operation to 2002 when damage of the main electrode occurred. The damage occurred during the noble metals washing operation by sodium carbonate. The exhausted air temperature from one of the main electrode was suddenly down and it meant air pass inside of the electrode was clogged by glass.

When the damage occurred in 2002, the first melter exchange project by the second melter, which was improved the structure of the bottom part, had been started because over five years have been passed since the start of the hot operation. So, JNC decided to give up operation by the damaged melter and accelerate the exchange project. After one and half year, JNC successfully finished the melter exchange project and restarted the facility operation on October of 2004. The vitrification campaign continued to the end of November and produced 20 canisters. The total number of produced canisters since the start of the hot operation in 1994 reached 150 canisters in total.

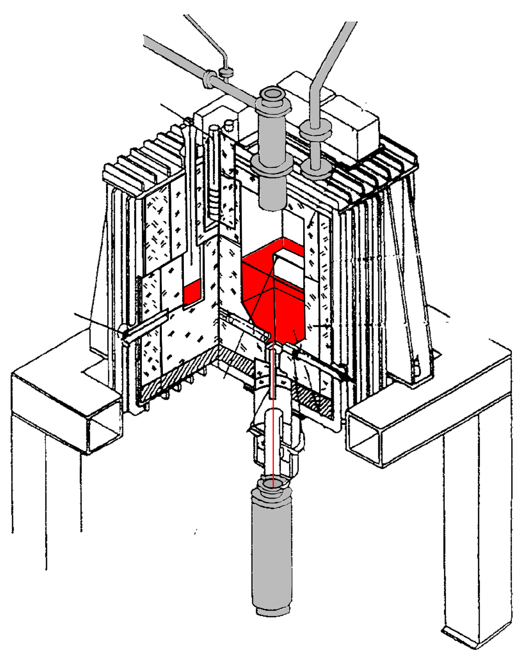


Fig. 2. The TVF melter

Melter Exchange

Manufacturing of the new melter started on April of 2003.

After completion of manufacturing the melter, the melter transferred to the Cold Mock-up Facility (CMUF) which located in the site of the JNC Tokai Works. The MUF is special facility designed for carrying out cold test of the melters for melter technology development. The facility is equipped with related equipment such as utilities supply, off-gas treatment system, canister handling system and so on. On May of 2003, the cold operation test of the second melter started and continued to the end of the December. After cold test, the melter transferred to TVF. On the other hand, in advance of the second melter transference, the first melter has removed from the original position to the dismantling area of the vitrification cell. After installing the new melter on the base frame, the third dimensional remote measuring was carried out to determine spatial relative position of jumper piping flanges. Based on the measured dimensional data, jumper tubes were manufactured and installed remotely. The melter installation work was finished on the end of August 2004.

To improve ability of draining for preventing noble metals accumulation on the melter bottom, new design of bottom structure was introduced for the second melter. Fig. 3 shows structure difference between the first melter and the second one. The first melter was employed step-up structure. The second melter attained flat slope by cutting the auxiliary electrodes heads that enable smooth movement of draining glass. The umbrella shape of the bottom electrode is also used for both purpose of getting continuous glass flow to the drain nozzle exit and blocking falling broken refractory. Air supply line for the bottom electrode is also employed to cool down as soon as possible after draining to prevent noble metals accumulation. The effect of the design change was checked in the cold operation test and no accumulation of noble metals was observed after draining all glass. In the cold operation test, operational parameters were fixed and parameters' differences from the first melter were confirmed. These new operational parameters were used to improve the operational manuals of TVF melter.

In the second melter operation, new electrode resistance observation technology is applied to detect noble metal accumulation in advance and also special glass removal device "mechanical chisel" was made. In the case of observing the resistance value of the main electrodes decrease less than the set up value, the melter operation is stopped for removing accumulated noble metals by the device.

VITRIFICATION TECHNOLOGY DEVELOPMENT

Melter Dismantling Technology

In the design of the TVF melter, corrosion of the electrodes and the refractory decides the lifetime of the melter because high temperature glass is very corrosive. So the melter is designed to be exchanged to new one every 500 pours and the old melter is dismantled. The generated solid wastes are divided into three kind of wastes; (1) glass separated from refractory and electrodes which will be contained in a canister and stored in TVF. (2) high radioactive solid waste which will be packed in hull canisters and stored in the HASW (High Active Solid Waste) Storage Facility in TRP. (3) LASW (Low Active Solid Waste) drums (200 L) stored in the LASW Storage Facility in TRP.

In the vitrification cell of TVF, there is special space for dismantling the melter remotely where the remote maintenance devices are equipped; the ceiling crane, the power manipulator system, the shielding window with a pair of the mechanical manipulator system and the turning table system (Fig. 4). This area is usually utilized for dismantling used tools, devices and piping for volume reduction.

TVF melter is composed of three main material; refractory, electrodes (Inconel), melter outer shell. The melter dismantling technology has been developing and special devices for dismantling were designed.
JNC has designed dismantling process and decided to use YAG laser for cutting the stainless outer shell and the

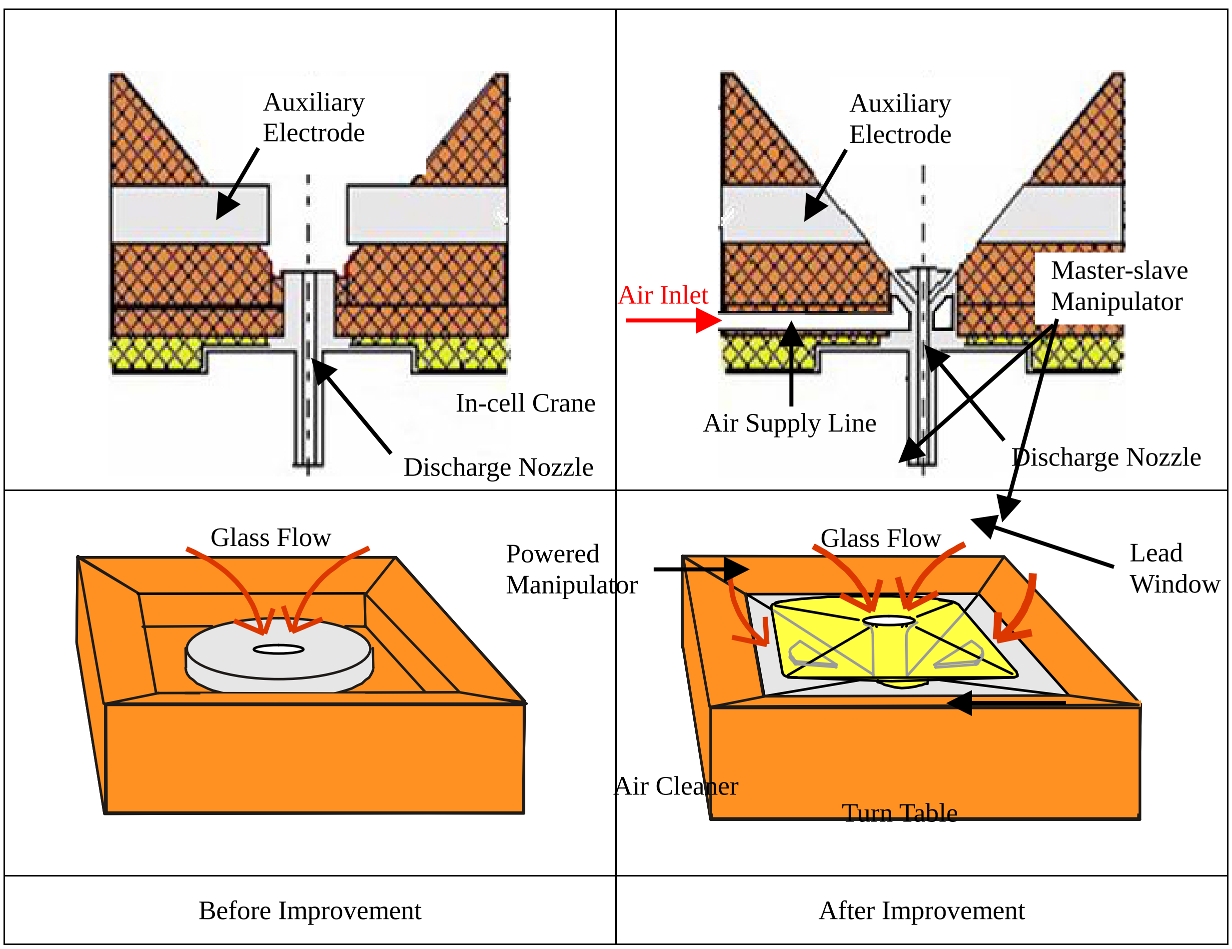


Fig. 3. Bottom structure improvement

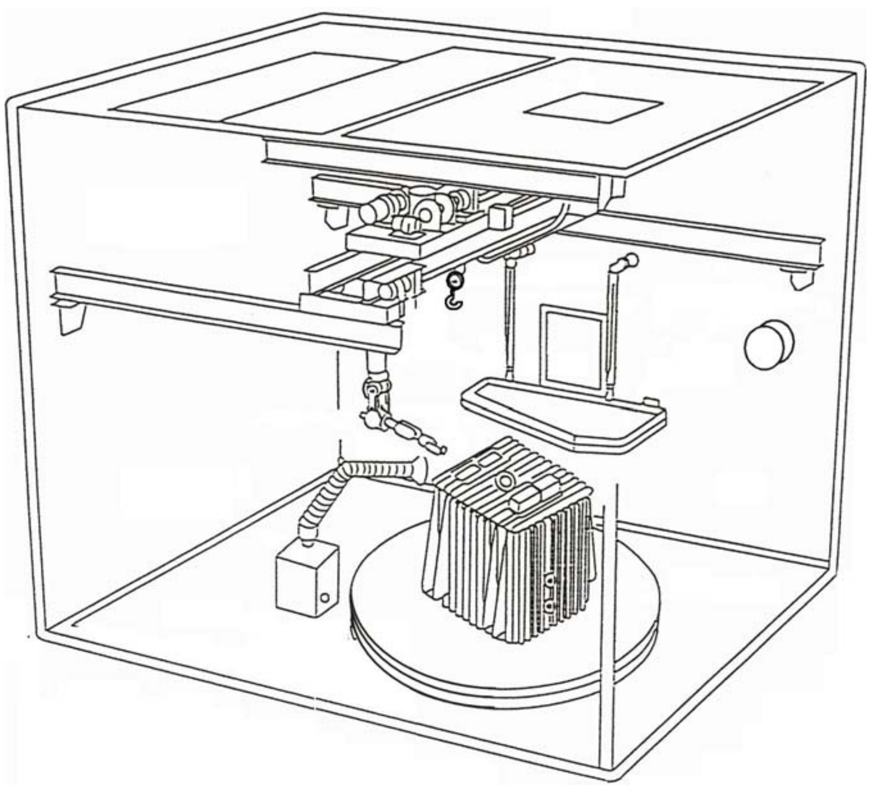


Fig. 4. Dismantling area in TVF

electrodes. Mechanical chisel is used for attached glass removal from refractory. YAG laser is also used for removing glass on the refractory, if necessary. Volume reduction of glass waste and HASW as much as possible is important issue for future disposal from the viewpoint of reducing natural burden and cost saving. So, separating technique of glass from refractory is a key technology. Decontamination technique of HASW is also important for reducing HASW volume. Data acquisition on degree and distribution of contamination is useful for next time dismantling of the melter to carry out more efficiently and inexpensively.

The melter dismantling is planned to start in 2005 and finish in 2007. Detail investigation of the damaged electrode will be done in the dismantling process and the damage cause will be finally resulted based on the observation data and taken sample data of the electrode.

Volume Reduction Technology

In Japan, the Final Disposal Act has already been enacted and the Japanese electric companies have already started payment into the waste disposal fund.

At present, the solidified glass includes about 25 % (weight percents) waste which composes 10% sodium dioxide and 15% metal elements (fission products, actinides etc.) If waste containment increased to 30 % (12 % sodium nitrate and

18 % metal elements), the number of necessary canisters decreases 23%. This means big cost saving for disposal. From this point, JNC has started the technology development of volume reduction.

When the glass volume is reduced, there is possibility of changing glass characteristics essential to the deep ground disposal. So, as the first step, JNC has been carrying out laboratory scale test of 30 % waste glass by measuring basic characteristics such as appearance, density, leaching rate, electric resistance and viscosity. As a result, so far, glass was homogeneous and satisfied the JNC's standard specifications of solidified glass.

Behavior of noble metals in the melter is another important issue to be confirmed, because there will be more possibility of noble metals' accumulation in the melter when much noble metals is fed to the melter. JNC plans to estimate the behavior of noble metals in the melter by the small scale melter test, the computer simulation and the full scale cold mock-up test.

Advanced Melter Design

As mentioned above, the melter is designed to be exchanged every five years because of electrode corrosion. Once the melter exchanged, the exchanging work interrupts the melter operation and dismantling work generates much waste which will become burden of nature in future. So to decrease the cost and the burden, there is necessary to develop an advanced melter which has longer life time and more robust characteristic. JNC feels such future needs strongly and decided to start design study for the advanced melter.

SOLIDIFICATION OF CONCENTRATED LOW RADIOACTIVE LIQUID WASTE

Facility Description

In TRP, following facilities were constructed adjacent to the Main Plant (MP) where main reprocessing process equipments are installed: the Auxiliary Active Facility (AAF), the Low Radioactive Liquid Waste Evaporation Facility (E), the Very Low Radioactive Liquid Waste Evaporation Facility (Z), the Oil Removal Facility (C) and so on. The LALW streams from MP are divided to five categories according to their activity and their generated process: medium radioactive liquid waste (MALW) ($3.7x10^4$-$3.7x10^5$ Bq/cc), low radioactive liquid waste (LALW) ($3.7x10^2$-$3.7x10^4$ Bq/cc), very low level liquid waste (VLALW) (<$3.7x10^{-2}$ Bq/cc), acid recovery distillate (3.7-3.7x10Bq/cc) and laundry waste ($3.7x10^2$ Bq/cc).

MALW is mainly generated in the shearing / dissolution off gas washing process, the used solvent treatment process and the analytical process in TRP. LALW comes mainly from the vessel off gas washing process and VLALW from the floor drain and the analytical process.

MALW and LALW are concentrated by the evaporator in AAF. VLALW and the acid recovery distillate are concentrated by the evaporator in the Z facility. The laundry waste is filtered in Z. The distillate of the evaporator in Z is transferred to C facility and oil is removed before sea release.

Concentrated liquid waste solution was transferred to the Bituminization Demonstration Facility (BDF) to solidify the waste by mixing with bitumen until 1997. BDF was constructed for processing concentrated liquid waste from TRP and started the hot operation in 1985. The building has four floors above the ground level and two floors of basement. On its first floor, there is the extruder cell in which the extruder is installed and the bitumen filling cell where drums are filled with bitumen-added liquid waste. The liquid waste adjusted its composition by adding reagents is fed to the steam heated extruder which dehydrates the liquid and kneads it with pure bitumen. The resulting mixture of waste liquid and bitumen is poured into empty drums.

OPERATION EXPERIENCE

JNC has operated the above waste treatment facilities safely and successfully and proved these facilities have enough decontamination ability to meet the strict sea discharge limits. In BDF, JNC processed 7,000 m^3 concentrated liquid waste solution and made about 30,000 drums of solidified products since 1982 to 1997.

But, unfortunately, the fire and explosion incident occurred on March 11 1997, at BDF. JNC took the restoration works to recover the confinement function of the facility and decontamination works. All the works completed by the beginning of September in 1998, and the safety function of the facility was recovered. After the incident, JNC decided not to use the facility for production of the bituminized product besides storage of the remaining concentrated liquid waste in the vessels of BDF. After decision to no more use of BDF for bituminization, JNC accelerated the construction project of LWTF and decided to build the new CLALW storage facility (LWSF) to cope with the shortage of storage capacity which is necessary during the construction time of LWTF. The construction of LWSF finished in 2002 and hot service started next year. LWSF has CLALW storage capacity of 1,500 m^3 in total.

LWTF CONSTRUCTION

LWTF has dual purposes to solidify CLALW and incinerate solid waste for waste volume reduction. JNC started the conceptual design in 1989 and finished the detail design in 1998. Construction of LWTF started in 2002 after getting legal approval on the safety design of LWTF by the Japanese authority. LWTF is under construction and JNC will start its hot operation in 2007.

LWTF has five floors above ground and two under ground floors. Main equipments for produce the ROBE solidified products are installed on the floors under ground and the incinerator is installed on the floor above ground.

On CLALW treatment process, JNC estimated that it had an economical advantage for future waste disposal. In LWRF, there is the coprecipitation / filtration process to divide CLALW flow into high radioactive slurry flow and low radioactive waste solution flow. In this process, radionuclides such as cesium and strontium move into the slurry flow.

Volume ratio of each flow is about 1:10. Each flow is fed to the evaporation process and solidified by mixing sodium borate. The capacity of the process is about 680 m^3/year and total volume reduction ratio is about 2.5. The ROBE product is made as interim form and additional treatment is necessary for final disposal, so JNC has started basic study on the additional treatment system which should be attached to the existing LWTF facility easily.

Type of the incinerator is a cylindrical furnace with cooling water jacket and it has capacity of 100 t/year. Total volume reduction ratio is about 15.

CONCLUSION

The first TVF melter started operation since 1995 and continued to 2002 when the damage of the main electrode occurred. In the hot operation, 130 canisters were produced.

The second melter was installed instead of the first melter on September 2004. The bottom structure design of the second melter was changed for smoothing draining which prevents accumulation of noble metals. The operation using the new melter started on October 2004 and finished successfully resulted in producing 20 canisters.

For dismantling the first melter, JNC designed and manufactured the YAG laser system. Installing these dismantling devices in TVF is under proceeding. The actual melter dismantling is planned to start in 2005 and finish in 2007. Detail investigation of the damaged electrode will be also done in the dismantling process.

JNC has started the technology development for volume reduction of the solidified glass. In the development, JNC tries to increase waste composition from 25 % to 30 % which will decrease a number of canisters 23 %.

After the fire and explosion incident of BDF, JNC decided not to use BDF for bituminization. Instead of BDF, LWTF is under construction and will be completed in 2006. In LWTF, the ROBE solidification process is employed to produce interim solidified product for decreasing the disposal cost.

REFERENCE

1. M. Yoshioka et al., "Vitrification Experience of TVF in Japan", Workshop of National Research Council, Commission Geosciences, Environment and Resources (1996)
2. Y. Senba et al., "Fully Remote Maintenance System in Tokai Vitrification Facility", Remote Techniques for Hazardous Environment, British Nuclear Energy Society (1999)
3. A. Aoshima et al., "Glass Melter Replacement and Melter Technology Development in the Tokai Vitrification Facility", Proceedings of 12th International Conference on Nuclear Engineering, April, 2004, USA