RECENT MAJOR RD AND D ACTIVITIES ON TRU WASTE MANAGEMENT

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ABSTRACT

Various kinds of TRU-bearing process wastes have been generated from MOX fuel fabrication and fuel reprocessing facilities at PNC. The nontreated (raw) waste from MOX fuel fabrication facilities has been successfully treated in Plutonium-contaminated Waste Treatment Facility (PWTF) since 1987. Combustible wastes and chlorinated organic wastes have been incinerated to be ash and then has been melted to be ceramic-like blocks. Metal wastes have been melted by electro-slag remelting. Approximately 75 tons (4,000 drums) of the raw Plutonium-contaminated Waste (PCW) have been reduced those volumes and weights to be 10 tons (45 drums) of ceramic blocks or metal ingots. The total volume reduction ratio is reached to approximately 1/100 at PWTF. Leaching rate of ceramic block is $1x10^{-5}$ g/cm^2 day (MCC-1 method). These operational result shows that the volume reduction and immobilization technologies for the raw PCW have been successfully demonstrated.

Concerning wastes from a fuel reprocessing, various kinds of low level raw wastes containing TRU nuclides are also generated.

Low-level liquid wastes containing high concentration of non-radioactive salt solution and spent solvent from the Tokai Reprocessing Plant (TRP) are incorporated into bitumen or epoxy resin, respectively. Solid raw wastes are packed in 200-liter drums or containers except that combustible waste has been incinerated. PNC has carried on R&D studies for volume reduction and radionuclide separation on low-level wastes to minimize conditioned TRU-bearing waste build up, and is planning to demonstrate the above developed technologies in Low-Level Waste Treatment Facility (LWTF). LWTF is now under design and will be constructed neighboring with the Tokai Reprocessing Plant.

INTRODUCTION

The technology development of TRU-bearing waste management is in progress concerning with wastes from plutonium/uranium mixed oxide (MOX) fuel fabrication facilities and LWR fuel reprocessing facility at the Tokai works, PNC. Approximately 110 tons of MOX fuels have been produced and 580 tons of spent LWR fuels have been reprocessed by December 1991.

Raw wastes from MOX fuel fabrication facilities have been processed in the PWTF since 1987. The reprocessing low-level liquid effluent is processed by evaporation, coagulation and neutralization prior to be released to the sea. A concentrated liquid waste containing high concentration of non-radioactive salts is incorporated into bitumen. A spent solvent is separated to dodecane and tri-butyl phosphate (TBP) using phosphoric acid. The TBP is incorporated into PVC or epoxy resin and separated dodecane is recycled to the TRP or incinerated. PNC has been developing alternative technologies to attain higher volume reduction by radionuclide separation *for the concent*rated liquid waste and the spent solvent.

So far, the treatment of raw process wastes such liquid as well as solid ones has been required to keep stable operation of upstream facilities such as fuel fabrication and reprocessing. Although, it is important in the sense of the waste management that the characteristics of waste to be disposed of should have suitable engineered barrier performance for candidate disposal system. Furthermore, it will be required to remove several radionuclides adequately from the waste to reduce the volume of waste to be conditioned for engineering storage on economical viewpoint.

STATUS OF TRU WASTE MANAGEMENT TECHNOLOGY AT PNC

TRU Bearing Wastes; Generation and Characteristics

Radioactive wastes containing transuranic nuclides generated from the operation of plutonium fuel fabrication facilities and the TRP are currently being safely stored in the storage facilities.

The solid raw PCW from MOX fuel fabrication facilities which are called PFDF, PFFF and PFPF are classified into three categories such as combustibles (paper, rags, wood, plastics, etc.), non-combustibles (metal, glass, concrete, etc.), and chlorinated organic materials (PVC, chloroprene, etc.). These wastes are stored packaged into 200-liter drums of 1.7m^3 steel containers. The volume constitution of the raw solid PCW are approximately 28% as combustible waste including filters, 44% as non-combustible waste, and 28% as other waste including chlorinated organic materials as shown in Fig. 1. The cumulative generation of the PCW has amounted to be approximately 4,000 m^3 by 1990 and 1,000 m^3 of which has been treated to be solidified product.

Since the active operation of the TRP was started on September 1977, about 580 tons of spent fuel have been reprocessed by December 1991. Meanwhile, radioactive wastes generated through the TRP operation is summarized in Table I.

The low-level liquid waste and the spent solvent are stored and incorporated into bitumen and epoxy resin, respectively. Then these solidified wastes are stored in the 200-liter drums at a storage facility.

Low-level solid wastes from the TRP are also classified into combustible, chlorinated organic, and non-combustible. The combustible waste has been treated at the incineration facility for volume reduction. The resultant incinerator ash

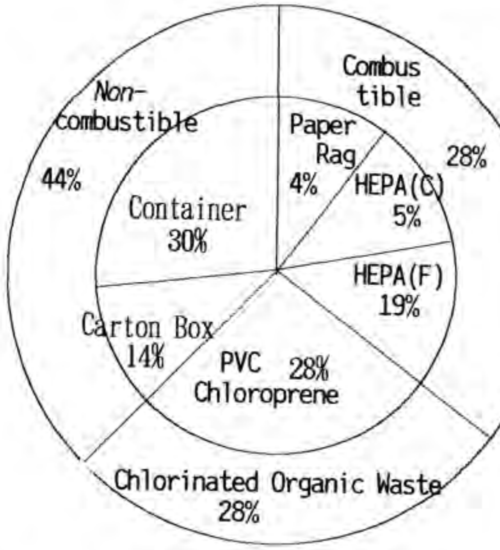

Fig. 1. Constitution of the raw PCW generated from MOX fuel fabrication facilities.

TABLE I

Low Level Raw Waste From the Operation of Tokai Reprocessing Plant

		TRU Contaminated Solid		
Type of Waste	Solid	Combustible	Non-combustible	Liquid
Source & Form	Leached hull Chopped endpiece Analytical labware	Paper, Cloths Wood, Rubber Plastics	Filter element Metal equipment	Second extraction U/Pu Purification Decontamination Solvent waste (TBP/Dodecane)
Package	Drum	Carton Box	Drum Concrete container	Storage vessel
Annual generating volume from 90 ton/y operation	~ 40m^3	~ 160m^3	~ 130m^3	~ 800m^3

and low-level solid raw wastes are packaged in 200-liter drums or multi-stage concrete container for storing in a storage facility.

Most of non-combustible solid raw wastes have been stored in the forms as they are generated and should be conditioned.

In order to evade the increase of storage cost, the volume reduction and immobilization technologies have been developed. Now development studies are still in progress also in considered with getting the final product optimized for waste disposal.

CURRENT TREATMENT OF PLUTONIUM CONTAMINATED WASTE (PCW) FROM MOX FUEL FABRICATION FACILITIES (1)

In November 1987, PNC has started the hot operation of the PWTF to condition the PCW wastes to be the durable waste forms such as ceramic blocks and metal ingots.

The objectives of PWTF are volume reduction of the process wastes generated from MOX fuel fabrication facilities as well as demonstration of the processes developed by PNC.

The PWTF has the throughput of 450m^3/y and can reduce the waste volume into one twentieth.

Each process unit has the throughput as shown in Table II.

Process Description

In the PWTF, the volume reduction and stabilization processes consist of the three kinds of operation unit: conventional incineration of paper, rags, wood, using fire grade controlled air incinerator and cyclone incineration of PVC, plastics and rubber; microwave melting for incinerated ash; Electroslag remelting (ESR) for metallic waste and incinerated ash. The incinerated ash can be melted without any additives to form 20-30kg ceramic blocks which are then placed in stainless steel canister. The major phases of the product are forsterite, anorthite and augite.

Metallic raw wastes are introduced into the ESR Unit and melted to form a slag/molten metal pool in a water cooled copper mould. The slag is a mixture of CaO, Al_2O_3 and B_2O_3. Contaminants such as plutonium and uranium oxide are converted into 100kg metallic ingots with little contamination at the bottom of the pool. After being used about twenty times, the slag material of the ESR is finally introduced to microwave melting by mixing with incinerated ash to form durable ceramic blocks. The incinerated ash is also added to the ESR as a part of the slag. Figure 2 shows a process diagram of the PWTF. Most processes are installed in the glove boxes and connected each other through connecting boxes.

Pretreatment and Sorting

The PCW in drum or container are introduced to the PWTF from the storage facility by transfer cart. After being assayed for plutonium content and surface contaminants, the wastes are charged into a glove box from the drum or container. These wastes, either in cartons or wrapped in PVC sheets, are sorted into four categories: metals, combustibles, HEPA filters and other wastes containing chlorine.

TABLE II

Throughput of each process

	(kg/d)
Incinerator	200
Cyclone Incineration	30
Electro-slag melter (Metal)	100
Microwave melter (ash)	30

The metal wastes are cut into 50mm x 50mm pieces with a shearing cutter and moved to the metal melting process by crane. Combustibles are cut and sealed in paper bags automatically and then sent to the conventional incineration process by gravity flow. HEPA filters are cut into several pieces with a circular saw and sent to the conventional incineration process by crane.

PVC and chloroprene wastes are cut into 5mm x 5mm pieces in two stages by shredding units and sent to the cyclone incineration process by gravity flow.

Conventional Incineration Process (1)

The conventional incineration process consists of a feeder, incinerator and off-gas treatment unit and has a throughput of 50kg/h. The incinerator is composed of a primary and a secondary combustion chamber and two kerosene burners. The calcination chamber is also equipped with an incinerator to calcine paints and tapes in metal wastes.

Bagged combustible wastes and pieces of HEPA filter are fed to the primary combustion chamber from the top of the incinerator through an airlock and adiabatic shutter. The wastes descend into the combustion zone where they are burned to ash with the two kerosene burners. The combustion gas and coarse dusts from the primary combustion chamber are carried over to the secondary chamber, filled with silicon carbide blocks for complete combustion.

The ash is removed from the bottom of the primary chamber and sent to the microwave melting process. The off-gas passes through a high temperature filter, diluter, HEPA filter, scrubber and mist separator (stainless steel mesh) prior to discharge from the stack to the atmosphere. Figure 3 shows the PWTF incineration process.

Cyclone incineration Process (1)

The cyclone incineration for organic chlorinated materials consists of a feeder, cyclone incinerator and off-gas treatment unit and has a throughput of 30 kg/d. The shredded wastes (5mm x 5mm) are fed through hopper and feeder to

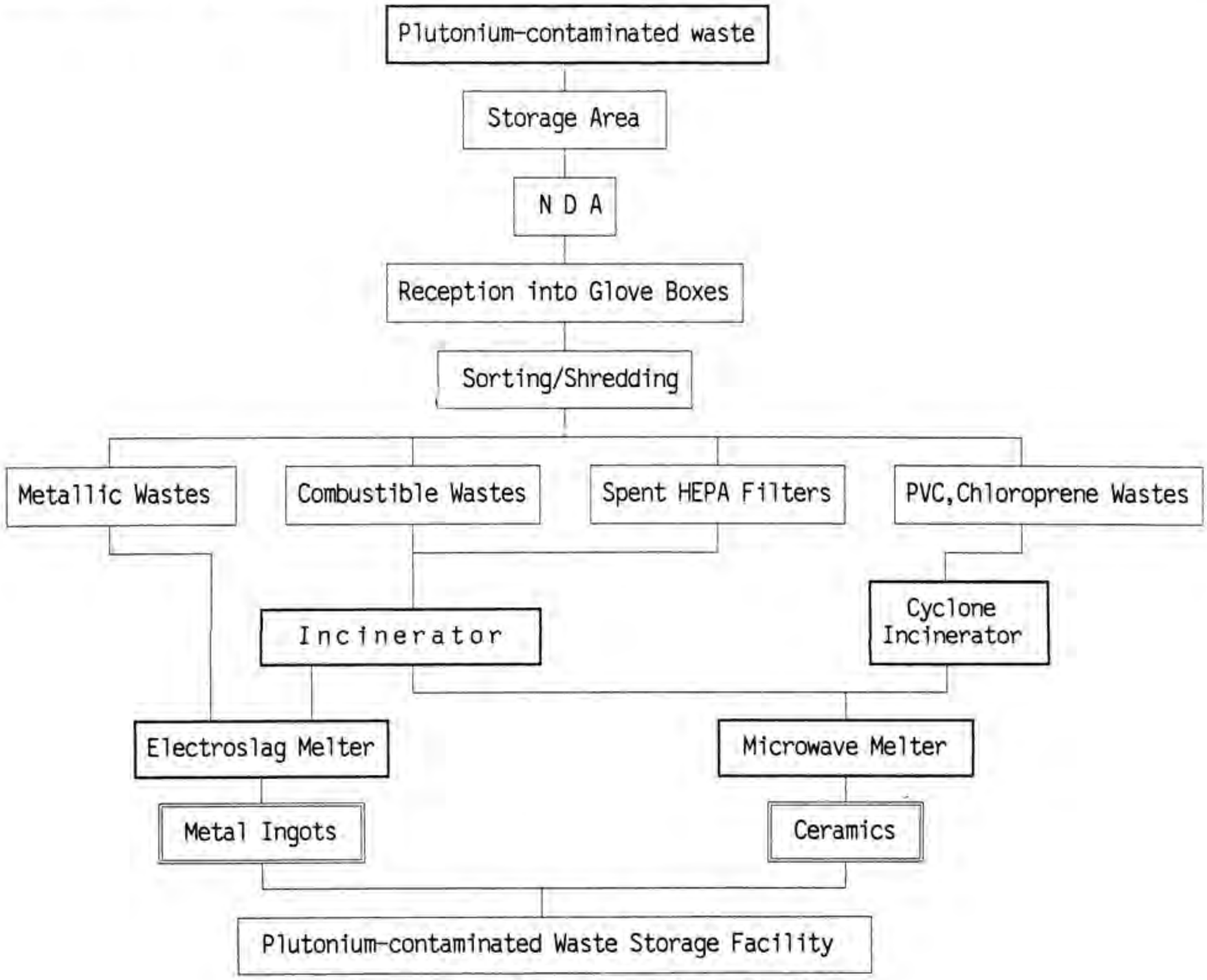

Fig. 2. Flow diagram of waste processing at the PWTF.

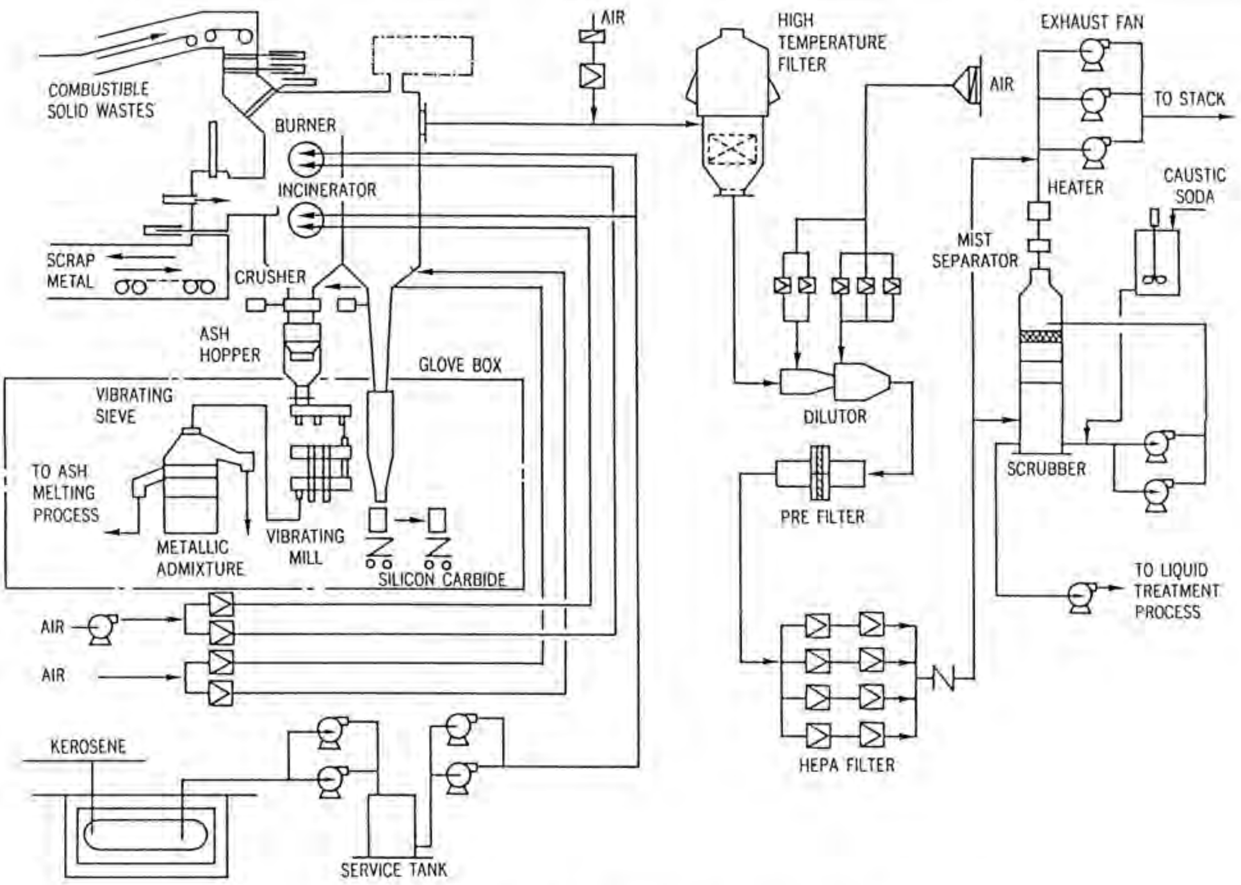

Fig. 3. Flow diagram of incineration process

the incinerator and circulate down in a spiral, forming ash at the bottom.

The cyclone incinerator is the cylindrical type of 56cm in diameter and 220cm in height.

The wall of the incinerator is made of high quality molded alumina. Ignition for the incineration is made by preheated oxygen rich air (30 vol% oxygen).

The ash is removed from the bottom of the incinerator. The off-gas is released from the stack through a cooling tower, partial condenser, HCl absorber, scrubber and HEPA filter.

Electroslag Remelting Process

The electroslag remelting (ESR) process consists of feeder, furnace, crusher and off-gas treatment unit and has a throughput of 100 kg/batch. The ESR furnace is composed of a mould, stool and tungsten electrodes. The water cooled mould is made of pure copper with inside dimensions of 220 mm x 380mm. About 100 kg of cut pieces of metal (50 mm x 50 mm) are fed into the molten slag pool at the rate of 60 kg/h.

To remove the metal ingot easily from the mould, the mould is designed to be divided into two parts. Two tungsten bars of 50 mm diameter are used as nonconsumable electrodes. Electric power is supplied to the tungsten electrodes through two copper bars aligned in parallel. Nitrogen gas is blown into the furnace to prevent oxidation of the electrodes.

To generate Joule heat in the slag, a stool arc method is used to start to melt the slag. The pieces of metal are gently fed into melted slag through a bucket. After melting, the slag is separated from the metal ingot and crushed for reuse.

Microwave melting Process (1)

The microwave melting process consists of a melter, waveguide and microwave generator and has a throughput of 5 kg/h. The melter consists of a tuner, crucible and isolator. The metal crucible is used as a primary canister. The size of the crucible is 130mm in diameter and 770 mm in height. The melter is installed in a glove box and microwaves are introduced through the waveguide from the generator installed outside the glove box. The generator can produce 10 kW microwave output with a frequency of 2,450 MHz.

Operating Experiences of the PCW Processing Technologies

During the four years operation of PWTF, approximately 69 tons of combustible wastes have been incinerated to be 6 tons of ash which has been melted to be ceramic block. The total volume reduction ratio of incineration and melting is approximately 100. Leaching rate of ceramic block is approximately 1 x 10^{-5}g/cm^{3}.day (MCC-1 method). Approximately 5 tons of chlorinated organic wastes have been burned by cyclone incineration and also 3.5 tons of metal waste have been melted by ESR. The volume reduction ratio of metal melting is approximately 6.

Table III shows the weight and volume reduction ratio of the incineration and melting.

TABLE III

Weight and Volume Reduction Ratio of the Incineration and Melting at PWTF (as of Nov. 1991)

Treatment Method Operational Results	Incineration	Ash Melting	Cyclone Incineration	Metal Melting
Treatment Waste	69 ton	6.3 ton (ash)	5 ton	72 (drums)
Product	6.3 ton (ash)	263 blocks (33 drums)	65 kg (ash)	12 (drums)
Weight Reduction Ratio	1/11	1	1/80	1
Volume Reduction Ratio	1/100		-	1/6

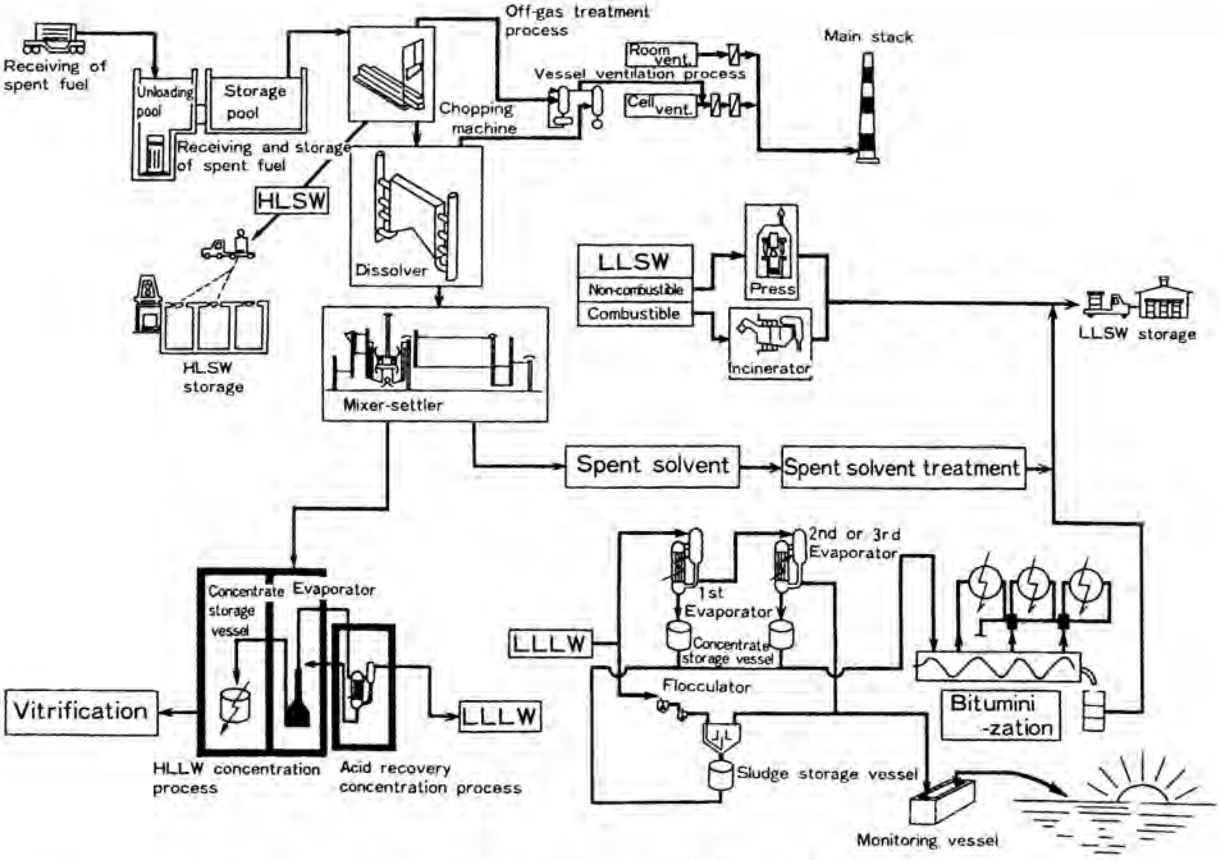

Fig. 4. Radioactive waste treatment in the Tokai Reprocessing Plant.

CURRENT TREATMENT OF TRU WASTES FROM THE TOKAI REPROCESSING PLANT (TRP)

The present treatment flow of reprocessing wastes is shown in Fig. 4. Along with the active operation of TRP, several waste treatment plants have been operated to support the reprocessing.

The Incineration Facility (IF) is for combustible waste, the Bituminization Demonstration Facility (BDF) is for the low-level concentrated liquid waste and the Solvent waste Treatment Facility (STF) is for spent solvent.

The IF has been operated since 1977 the design capacity of which is 400 kg/day. The new incineration facility has constructed and just started in 1992 as the replacement of the former one. The new facility is installed two types of incinerators such as a conventional incinerator same type as PWTF for miscellaneous combustible waste and a solvent incinerator for dodecane and charcoals. The throughput of the conventional incinerator is 500 kg/day, and that of the solvent incinerator is 100 kg/day.

The BDF has been started operation in 1982 to demonstrate a solidification technique with bitumen. This plant utilizes a horizontal extruder as bituminization process. The concentrated liquid waste from the TRP contains relatively high inactive nitrate salt as it is received in the bituminization facility.

The concentrate separated at the evaporation and flocculation processes at the TRP are received in the BDF. The barium and ferrocianide salts are added for the fixation of strontium and cesium, respectively, and silver nitrate is added for iodine.

The pretreated waste is transferred into the extruder. In the extruder, the liquid waste is blended with bitumen, dehydrated and poured into drums. The dehydration capacity of extruder is about 200 liters/h. The condensate from extruder is returned to the liquid waste treatment process in the TRP. The volume ratio of the feed liquid to the bitumen product is approximately 1/1.

Now, about 20,000 drums of bituminized wastes are produced and stored.

The spent solvent received in the STF is washed with sodium carbonate at first, followed by separation of dodecane and TBP using phosphoric acid and pure water. After removing impurities by silicagels, purified dodecane is either reused in the reprocessing process or burnt with incinerator. The separated TBP is mixed with PVC or epoxy resin in drums for uniformalizing, and the resultant solidified waste are in storage.

Since hot operation started in 1986, spent solvent about 260 m^3 has been treated until the end of 1991. About 50 m^3 of purified dodecane has been recycled and solidified product of 1,100 drums of 100-liter drum are now in storage.

INNOVATIVE TECHNOLOGIES ON TRU WASTE CONDITIONING (2)

In order to advance the TRU waste management technology, PNC has been developing the innovative technologies for conditioning the TRU wastes.

The viewpoints of the technology development are as follows:

1. To get the higher volume reduction to minimize the final waste product for engineering storage and disposal.
2. To make stable waste forms for easiness in handling and storage as well as avoiding the sophisticated performance assessment for waste isolation.
3. To avoid the presence of organic material in the radioactive wastes.

Some increase of treatment steps should be allowed in considering with these advantages in looking at the overall waste management.

Radionuclides Removal From the Low-Level Liquid Waste (2)

The radionuclides removal technology for the low-level concentrated liquid waste containing sodium nitrate arising from the TRP is being developed to be replaced with bituminization technology to get higher volume reduction. Almost of radionuclides are separated from liquid stream and solidified into extremely small volume and most part of the waste stream can be released to the sea or recycled. The process mainly consists of precipitation, removal of carbonic acid, co-precipitation/ultra-filtration and ion-exchange units. The process concept is shown in Fig. 5.

Iodine is separated from the liquid waste by the precipitation with silver nitrate and ultra-filtration at the pH 8. The precipitation method has been demonstrated using actual waste at laboratory scale.

Uranium forms a soluble uranyl-carbonate with carbonic acid ion. Because it is difficult to separate uranyl-carbonate by the co-precipitation and ultra-filtration, it is necessary to removal of carbonic acid ion before the co-precipitation. Uranyl-carbonate is converted to uranyl-nitrate by adjustment pH 2 adding nitric acid to the waste. Carbonic ion is driven out from the waste in the form of carbon dioxide by heating and air-blowing at pH 2. The engineering scale experiments on co-precipitation and ultra-filtration using simulated waste showed that uranium was effectively separated in the condition of pH 8 to 10, 60°C and 100 ppm co-precipitant concentration and using a hollow fiber ultra-filter made of poly-sulfonic acid. The uranium concentration in the filtrate was decreased in proportion to the co-precipitant concentration, and was below the sea discharged level (2.96×10^{-2} Bq/cm^3) at 100 ppm of co-precipitant concentration. The relation between uranium concentration in filtrate from ultra-filter and co-precipitant or ferric ion concentration is shown in Fig. 6.

Ruthenium, cerium and zirconium are also separated effectively at the same condition as that of the uranium separation.

Cesium and strontium are separated by cobalt ferrocyanide ion-exchanger and sodium titanate ion-exchanger at the pH 7, respectively.

The laboratory-scale hot experiment using the actual very low-level liquid waste (10^1 Bq/cm^3) showed the radioactivity was reduced below the sea discharged level.

The volume reduction ratio of the low-level liquid waste treatment process is expected to be about 1/50 as compared with that of the present bituminization.

Decomposition and Radionuclides Removal of the Spent Solvent (2)

The radionuclides separation technology for the spent solvent arising from the TRP has been developed to mineralize and classify the wastes in TRU and non-TRU. The process mainly consists of decomposition of spent solvent and co-precipitation units. The process flow is shown in Fig. 7.

It is shown by using simulated spent solvent that the spent solvent was effectively decomposed to inorganic phosphoric acid by hydrogen peroxide and copper oxide catalyst in the condition of 100°C, 1 atm.

Plutonium co-precipitation and separation tests for phosphoric acid solution as the simulated liquid waste of decomposed solvent were conducted using lanthanum, zirconium, iron and calcium as a co-precipitant.

The fundamental tests showed plutonium was effectively separated in the condition of pH 4 to 8 and lanthanum concentration of 10,000 ppm. The volume reduction ratio of the spent solvent treatment process is expected to be 1/20 instead of the conventional plastic incorporation of 1/1.

Decontamination Techniques (3)

Decontamination techniques are classified into two groups, namely "primary decontamination techniques" and "complete decontamination techniques".

The former envisages the removal of the loose contaminants to reduce the exposure dose rate involved in handling nuclides and to prevent the spread of contamination mostly considering the operation of the facilities.

The latter aims at the absolute reduction of radioactivity down to the background level for minimizing the amount of the wastes.

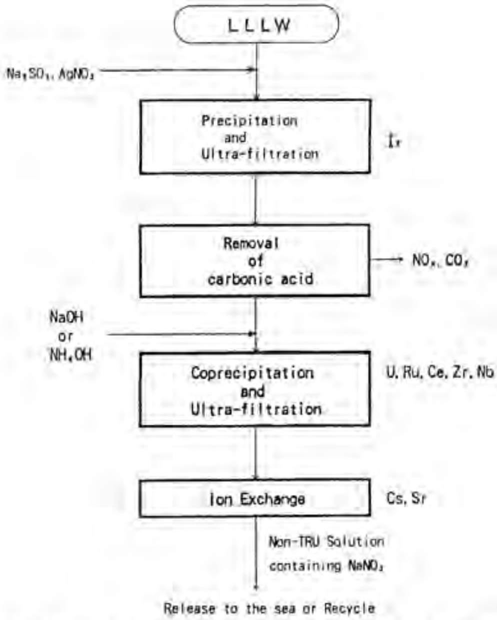

Fig. 5. Radionuclides removal process for the low-level liquid waste (2).

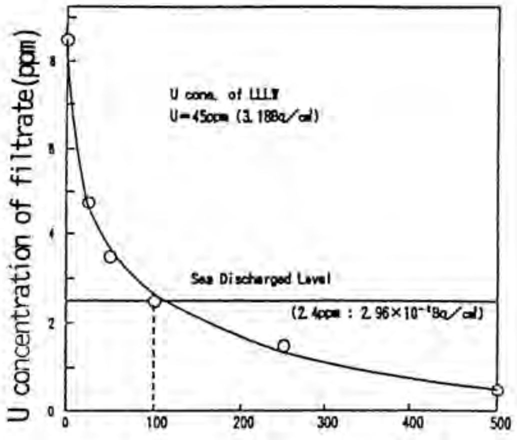

Fig. 6. The relation between uranium concentration filtrate and co-precipitant (Fe) concentration.

PNC is now undertaking the development of some complete decontamination techniques as well as the improvement of the conventional decontamination techniques to apply for TRU-contaminated wastes from reprocessing and MOX fuel fabrication facilities. The technology development is also related with the wastes from decommissioning. The overall technology development plan and objectives are shown in Fig. 8.

The techniques under development are ice blasting, electropolishing, and REDOX decontamination. The application of the decontamination by electroslag-remelting on the reprocessing wastes is also in consideration.

The decontamination system based on the current development will also be equipped in the LWTF to decontaminate the noncombustible TRU raw wastes into non-TRU management and also to get higher volume reduction.

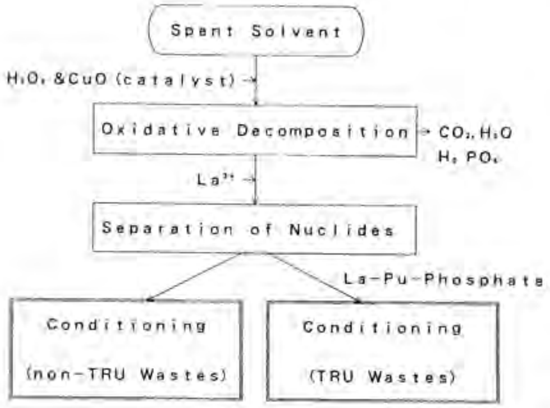

Fig. 7. Decomposition & radionuclides removal process for the spent solvent (2)

CONCLUSION

Advanced technology development is in progress to get higher volume reduction and radionuclide separation as well as to get the stable waste products.

1. TRU waste from MOX fuel fabrication facilities have been successfully treated and conditioned in plant scale at PWTF.
2. The radionuclide separation processes have been developed for low-level concentrated liquid waste containing sodium nitrate and spent solvent.
3. The decontamination technology development for TRU-contaminated solid waste has been carried out to get more effective method aiming at the absolute reduction of contamination down to background level.
4. LWTF for miscellaneous low-level solid waste from reprocessing plant as well as nuclides removal from low-level liquid waste with high sodium nitrate salt content is planning to construct to demonstrate the advanced treatment process for the reprocessing wastes.

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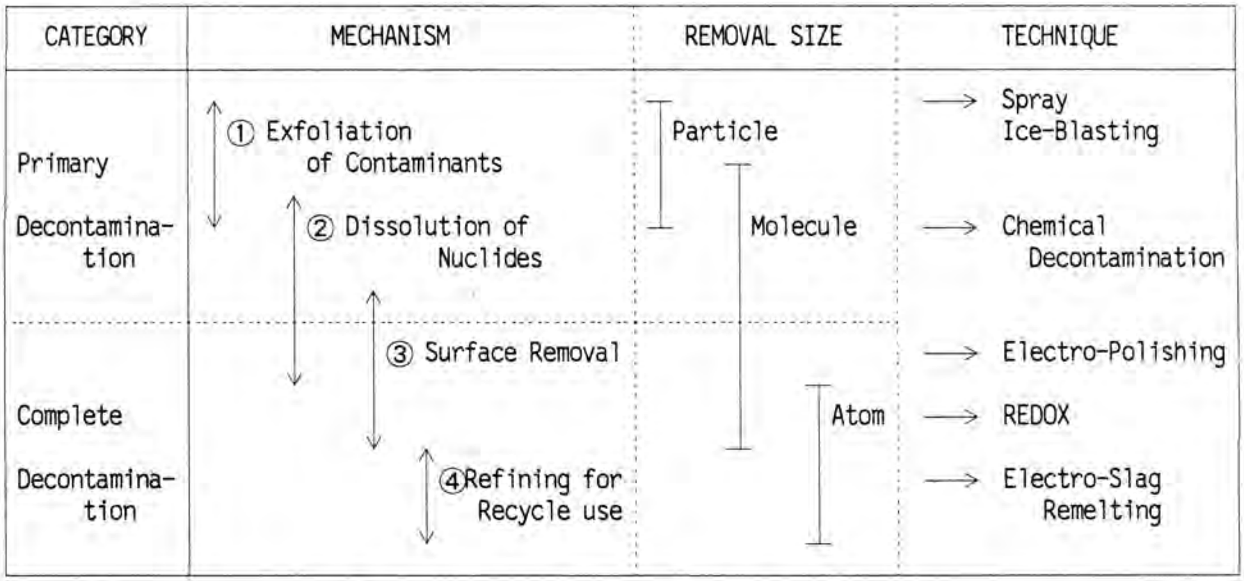

Fig. 8. Decontamination techniques and objectives.