THE INTRODUCTION OF NEW WASTE TREATMENT SYSTEM AT THE FUGEN NUCLEAR POWER STATION

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ABSTRACT

Advanced Thermal Reactor Fugen Nuclear Power Station is planning to introduce "spent resin volume reduction and stabilization processing device" and "laundry drain filtering device" in radioactive waster treatment system. As for the former device, we demonstrated sufficient performance of this device by carrying out the confirmation test using real spent resin relating to the following points: performance of volume and weight reduction; change to inorganic material; detoxification; and retention and transfer of nuclide, etc. As for the latter device, we are studying two methods now. However, we confirmed that it is possible to apply the removal performance of radioactivity and Chemical Oxygen Demand to real machine by carrying out hot test.

INTRODUCTION

The Advanced Thermal Reactor Fugen Nuclear Power Station (hereafter, referred to as, "Fugen") is a heavy water moderated, light water cooled, pressure tube type reactor aimed at diversifying and effectively using nuclear materials for the energy security in Japan. Fugen started operation in 1979 as a prototype reactor (electric power: 165MWe). Although the plan of demonstration reactor was supposed to advance to the next step, it was cancelled due to the economic issues. Fugen was shutdown permanently in 2003 and completed the mission to develop an ATR.

From now on we have to develop several technologies not only for decommissioning but also to reduce the amount of solid waste for disposal. Therefore, we are planning to install new treatment systems for solid and liquid radioactive waste.

Solid Waste Treatment System

Over the past 24 years, approximately 220m^3 of spent resin was generated and stored at Fugen. It is necessary to change the spent resin from organic to inorganic material in order to ensure its long-term stability in the disposal site after solidification. We are considering the introduction of the processing device at Fugen to reduce the volume of spent resin and stabilize it using low-pressure oxygen heated and activated by a plasma .

Liquid Waste Treatment System

We are planning to replace the laundry dry cleaning system that uses chlorofluorocarbon (CFC) with a wet wash system to cease CFC use and protect the environment. Two filtration processes are being studied to treat laundry liquid. The difference between two methods is in the catalyst that is used for decomposition of organic material in the waste water. One method decomposes organic material with microorganisms, and the other method uses ozone.

EXPERIMENTS

We carried out several tests on the following technologies: "spent resin volume reduction and stabilization process" and "laundry drain process."

Technology of Spent Resin Volume Reduction and Stabilization Process

Principle/Characteristics.

This device is called "low-pressure oxygen plasma method." (See Figure 1) This device will reduce the volume and stabilize spent resin. It uses heated and activated oxygen produced by the plasma chemistry created by injecting oxygen into a low-pressure atmosphere. In addition, the inductively-coupled plasma (ICP) method is employed to generate the plasma. This method is based on the principle that electrodeless discharge is generated by the action of an inductive electric field when applying high frequency current to the inductive coil which is installed in plasma production area. This method has the advantage that it is possible to simplify the structure, reduce the damage of components by plasma, maintain the stable plasma.

Process of Treatment.

For anionic resin, the ion-exchange group is generally started decomposing at 120 to 310°C depending on the nature of ion-exchange resin and base material of resin is decomposed at approximately 500°C. For the cationic resin, the ion-exchange group is generally decomposed at the 370°C and base material of resin is decomposed at approximately 700°C. The process of treatment has two steps based on the characteristics of resin.

First Treatment (Low Volume Reduction Process). Pyrolysis gas, which is generated by heating the spent resin at 400°C, is oxidized and decomposed by using the heated and activated oxygen caused by plasma. This method allows resin component to reduced its volume and weight to approximately 1/4 or 1/5.

Second treatment (High Volume Reduction Process). After the first treatment, the resin is heated at approximately 700°C and then is oxidized and decomposed by direct contact with heated and activated oxygen plasma. If this treatment is continued, the volume and weight of this resin component can be reduced to less than 1/20.

The resin component, which is composed of carbon and oxygen through the above treatment process, is oxidized and decomposed to CO_2, CO and H_2O. The functional group (such as SO_3, NH_3 etc.), which is capable of ion-exchange, is reduced to SOx and NOx and then exhausted as gas. Ion-exchange resin is reduced in volume and weight while enhancing carbonization by pyrolysis as well as direct oxidization. Metal ions, which are adsorbed into ion-exchange resin, remain as metal oxide and sulfide etc. in the residue after treatment. Therefore, spent resin volume and weight is reduced and changed to carbonized resin. In addition, radioactive material in the spent resin, is capable of recovery together with carbonized resin because it is changed to an oxide.

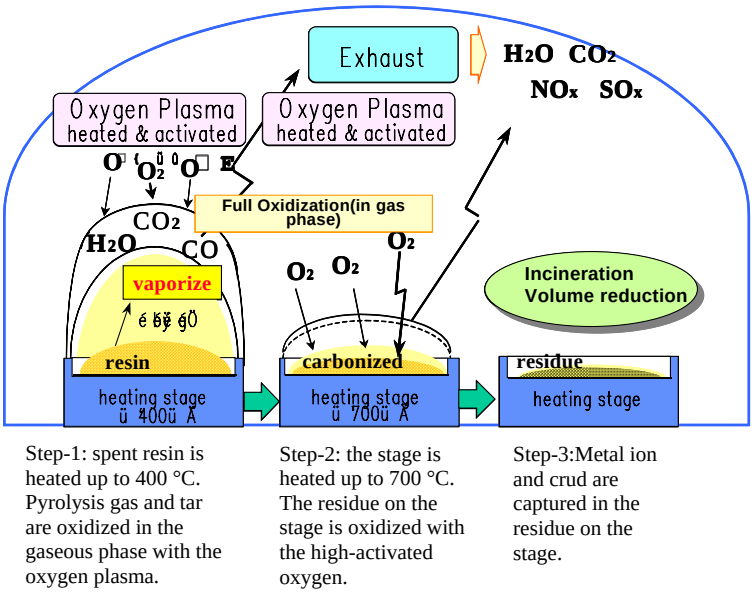


Fig. 1. Concept of Low-Pressure Oxygen Plasma Method

Performance Confirmation Test.

Performance tests were carried out with large and small test devices in accordance with the following items simulated and actual spent resin. The outline of small test device is shown in Figure 2 as an example of test device.

- Volume and weight reduction performance
- Conversion to inorganic material performance
- Detoxification (EDTA, hexavalent chromium) performance
- Retention and transfer of nuclide (Co-60, C-14, H-3) performance

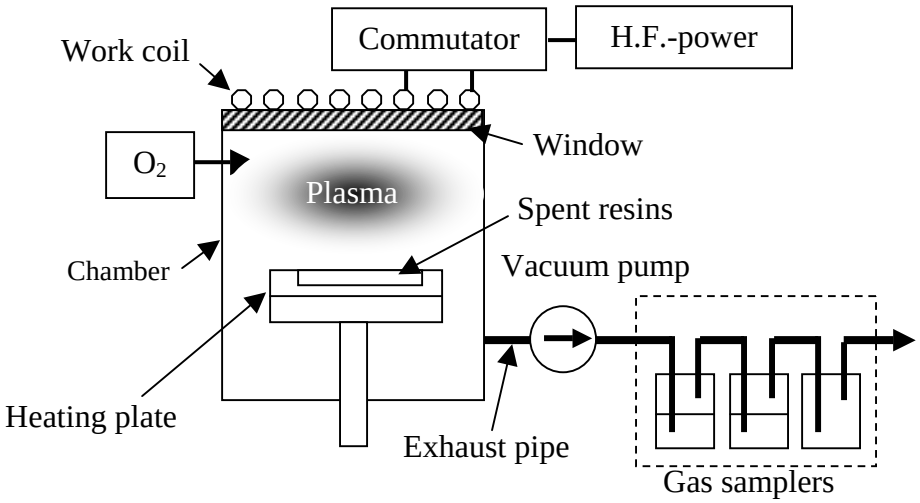


Fig. 2. Outline of small test device

Confirmation of Volume and Weight Reduction Performance

Method of Test. Volume and weight reduction of two actual granulated spent resins (condensate water purification system, heavy water purification system) and one powdered spent resin (pool water purification system) were verified by operating small test device for 210 minutes including 1 hour as the first treatment, 2 hours as the second treatment and a warm-up period. In addition, these results were confirmed according to the steps divided into the first treatment alone and both the first and second treatments.

Test Results. The results of the test are shown in Table I.

- We determined that the volume and weight of the spent resin were reduced 1/3 respectively after the first treatment alone.
- We determined that the volume and weight of the spent resin were reduced 1/10 respectively after the first and second treatments.
- Both reduction rates of powdered spent resin were higher than bead resin .
- The weight reduction rate was higher than volume reduction rate.

Table I. Test Results of Volume and Weight Reduction

Evaluation items	Treatment step	Condensate water purification SR (Granulated SR)	Heavy water purification SR (Granulated SR)	Pool water purification system SR (Powdered SR)
Volume reduction rate	1st	66	68	89
	1st+2nd	90	93	98
Weight reduction rate	1st	68	75	89
	1st+2nd	96	98	99

Unit: % (This table is based on the calculation results)

Conversion to Inorganic material Performance

Method of Test. Principal component of resin consists of C (Carbon), H (Hydrogen), N (Nitrogen) and S (Sulfur). The conversion of resin to inorganic material was confirmed for this device. Conversion to inorganic material was verified by treating two simulated spent resins (in heavy water purification system, chemical decontamination system) having different componential rates.

Test Results. The results of the weight reduction are shown in Figure 3. Crud and metal ions adsorbed into resin was indicated as "Other" in Figure 3. We determined that the weight reduction rate was 99% confirming good conversion of resin to inorganic material. As for the chemical decontamination spent resin , the weight reduction rate was only 92% because of remaining metal ions and crud.

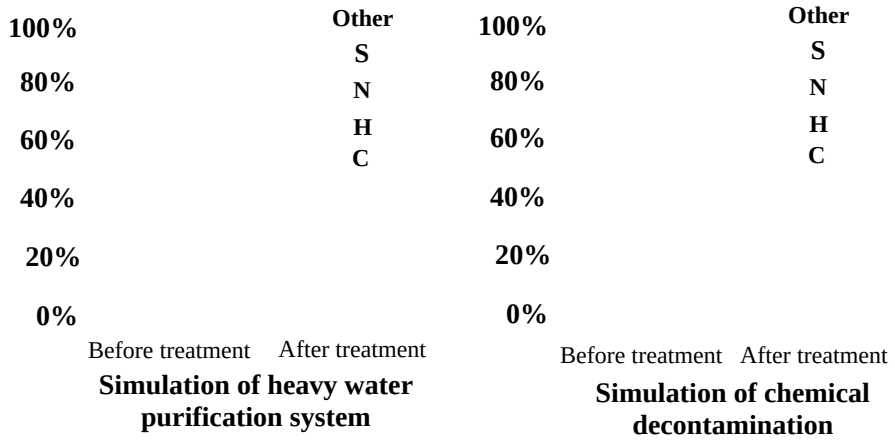


Fig. 3. Test Results of Performance of Change to Inorganic Material

Detoxification Performance

Method of Test. Spent resin used for system chemical decontamination at Fugen, contains harmful chelate (EDTA) and hexavalent chromium that must be detoxified prior to disposal. Detoxification was verified by treatment of the chemical decontamination spent resin, in a small test device.

Test Results. We determined that EDTA was reduced from 1360 mg/kg to below the detection limit (<15mg/kg) after the first treatment. In addition, we determined that there was 0.02mg/l of hexavalent it of 1.5mg/l for industri

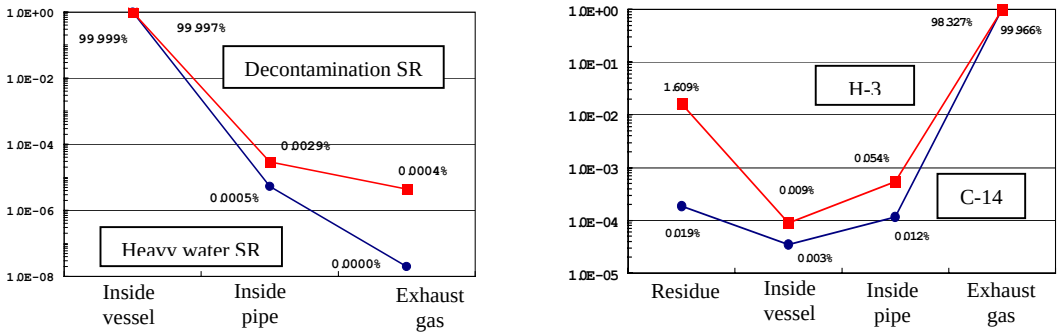
Retention a

Method of Fugen, the particulate o prevent of

radioactive release . In addition, C-14 and H-3 will be discharged, after dilution, and avoid being contained in the solidified waste to be buried. Two kinds of spent resin (heavy water purification system and chemical decontamination system) were treated by the small test device to determine the retention of Co-60. The release of C-14 and H-3, was determined from actual heavy water purification system spent resin.

Test Results. The results of the test are shown in Figure 4.

- Release of Co-60 (metal nuclide) to the exhaust system was below 10E-4, and its retention rate in the waste container was more than 99.99%.
- Release rate of C-14 to the exhaust system was more than 99.9%, and its retention rate in the waste container was less than 0.1%.
- Release rate of H-3 to the exhaust system was more than 98%, and its retention rate in the waste container was less than 2%.



Co-60 **C-14 and H-3**

Fig. 4. Retention and Transfer of Nuclide Performance

Technology of Laundry Drain Process (Membrane Separation Activated Sludge Process)

Principle/Characteristics.

The activated sludge process is a method to purify drain liquids by the metabolic function of activated sludge (microbial community) composed mainly of bacteria, protozoan and metazoan. Organic material in waste water is oxidized and decomposed by microorganisms to carbon dioxide and water in an aerating tank (septic tank). This method has already been employed widely in general industry. In general, treated water including sludge is transferred to a sedimentation tank and separated into solid and liquid by naturally settling out after aeration. However, this method requires a large sedimentation tank and there is insufficient space at the site. In order to downsize the processing device, we are planning to employ "membrane separation activated sludge process" which uses a membrane in the aerating tank for the solid-liquid separation instead of a sedimentation tank. The advantage of this method is that it is possible to maintain a higher concentration of sludge in the aerating tank . In addition, it is possible to completely remove the suspected solids in treated water by membrane filtration. The process with the septic tank is shown in Figure 5. Laundry drain, which is received in the

sedimentation tank, is oxidized and decomposed by the action of activated sludge. The treated water is transferred to the treated water tank through the separation membrane (pore size: 0.4μm). In addition, the sludge, which remains in the septic tank, is concentrated with sludge thickening equipment and collected. Finally, it is burned in an incinerator.

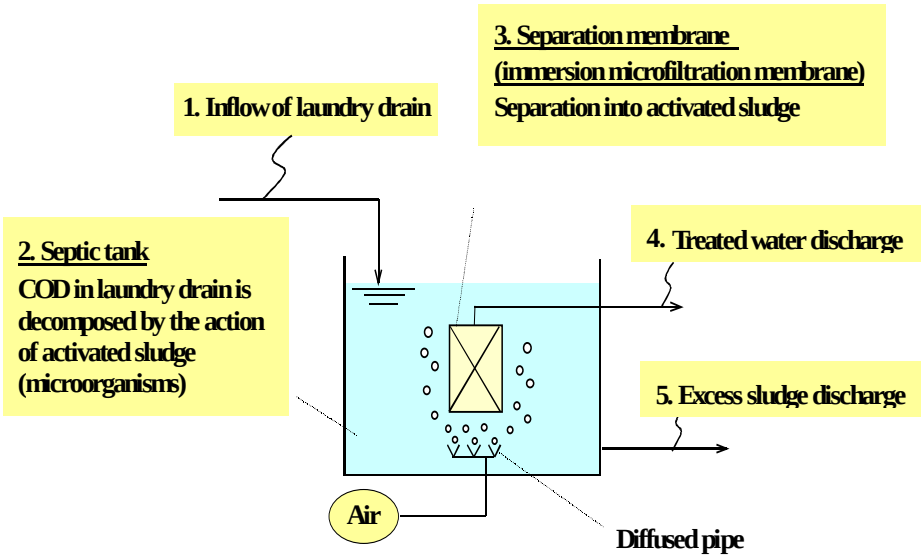


Fig. 5. Outline of Treatment Method (Activated Sludge Process)

Cold Test

Effects of the Concentration of Activated Sludge on Co Adsorption. The purpose of this test is to research the effects of the concentration of activated sludge on Co adsorption of microorganisms. The simulated water, which was injected with 1mg/l of Co in 400ml of purified water, was treated in three septic tanks differing in sludge density (3000, 7000, 12000mg/l) for 24 hours. After passing through the filtration membrane (0.45μm), the treated water was collected. This experiment shows that Co concentration in treated water after treatment is 0.067, 0.056, 0.039mg/l at each sludge density and DF (Decontamination Factor) is 15, 18, 26. We determined that absorption and DF was improved by increasing activated sludge density.

Relationship of Co Concentration Between Before and After the Treatment. This test investigated the effects of Co concentration in waste water on DF. We studied Co concentration after treatment of three concentrations (0.5, 1.0, 2mg/l) in the septic tank (sludge density 7000mg/l) for 24 hours (including treated water with separation membrane). From this test, we confirmed that Co concentration after each treatment was 0.03, 0.07, 0.13, and DF was approximately 15

constantly. Therefore, we found that DF was only dependant on the concentration of active sludge.

Effect of Detergent on Co Adsorption.

The purpose of this test is to research whether Co components in waste waster will be difficult to adsorb with microorganisms after combination with detergent. First, we prepared three kinds of simulated water (Co concentration is 1mg/l before the treatment) and added 0.1% of several kinds of detergent into these simulated laundry water. Finally, we treated these simulated waste in the septic tank (sludge density 7000mg/l) for 24 hours and passed them through the filtration membrane (0.45μm). We compared Co concentration of these simulated wastes treated by the procedure above with one free of detergent. From the results of this test we recognized that there was some impaired adsorption for two kinds of detergent because the Co concentration I was approximately 1.7 times higher than the case of detergent free waste.

Test of Soluble Co Treatment with Sodium Hypochlorite

I mentioned that adsorbability of soluble Co to activated sludge declined due to the coexistence of detergent. Test of soluble Co treatment with sodium hypochlorite is for insolubilizing soluble Co and oxidizing and decomposing detergent components as a pretreatment to avoid declining performance. Firstly, we added sodium hypochlorite (0, 5, 25, 50, 250, 500mg/l) into simulated water (Co concentration=1.0mg/l) and agitated it for 10 minutes. After passing through filtration membrane (0.45μm), we checked Co concentration of this water. From the results of this test we determined that the Co concentration of the treated water reduced noticeably when adding more than 250mg/l of sodium hypochlorite. The DF was 14 when adding 500 mg/l. In addition, Co concentration of treated water was 0.62mg/l (DF=1.6) when adding 5mg/l sodium hypochlorite, however, its Co concentration changed to 0.05mg/l (DF=19) by extending the treating time from 10 minutes to 12 hours.

Hot Test: Verification Test of effect of pretreatment with sodium hypochlorite

Method of Test. The purpose of this test I was to verify a relationship between sodium hypochlorite and Co concentration of treated water and change of filtration rate by using four kinds of test water based on actual laundry drains as shown in Table II. We added sodium hypochlorite (given concentration: 0, 5, 250, 500mg/l) into test water (400ml), agitated it for 2 hours, and passed it through the filtration membrane (0.45μm).

- Test water 1: Washing five (low-level) contaminated uniforms with 30 l of water containing 30g of detergent, and rinsing with 160 l of water. Then half of laundry drain and rising one are mixed with water to 120 l. Finally adding 140ml of concentrated waste water into these mixed liquids in order to make 5-10Bq/ml radioactive concentration of test water.
- Test water 2: Washing four uncontaminated uniforms with 20 l of water, adding 2g of detergent into its laundry drain and also adding some crud to make 5-10Bq/ml radioactive concentration of test water.

- Test water 3: Washing five (low-level) contaminated uniforms with 20 l of water containing 2g of detergent, and adding some crud to make 5-10Bq/ml radioactive concentration of test water.
- Test water 4: Washing four uncontaminated uniforms with 20 l of water, adding 2g of detergent into its laundry drain and also adding slightly concentrated waste water to make 5-10Bq/ml radioactive concentration of test water.

Table II. State of Test Water.

Test water No.	Co Concentration (Bq/ml)	COD (mg/l)	SS (mg/l)	Electric conductivity (mS/cm)
Test water 1	8.49E+0	54	96	0.92
Test water 2	8.40E+0	13	20	0.15
Test water 3	7.02E+0	58	32	0.30
Test water 4	9.44 E+0	27	174	1.70

COD: Chemical Oxygen Demand
SS: Suspended Solid

Results of Test. The results of the test are compiled in Table III.

- As for relationship between sodium hypochlorite and Co concentration, we determined that for the case of test water 2 and 4 the effect doubled (i.e., Co encapsulation), however, using test water 1 and 3 had no effect. We think there was a possibility of a different Co ion configuration in the hot wastewater test.
- We determined that the filtration rate increased 1.5-1.8 times by adding 50mg/l of sodium hypochlorite.

Table III. Results of Drain Treatment Test

Test water No.	Hypochlorous Na (mg/l)	Co concentration (Bq/ml)	DF	Filtration rate (ml/sec)
Test water 1	0	8.23 E-2	103	0.26
	5	8.85 E-2	96	-
	250	9.36 E-2	91	0.48
	500	9.18 E-2	92	-
Test water 2	0	5.50 E-2	153	-
	5	3.36 E-2	250	-
	250	3.31 E-2	254	-
	500	3.61 E-2	233	-
Test water 3	0	9.76 E-2	72	0.32
	5	1.01 E-1	70	-
	250	1.02 E-1	69	0.48
	500	9.56 E-2	73	-
Test water 4	0	8.32 E-2	113	-
	5	5.53 E-2	171	-
	250	4.74 E-2	199	-
	500	3.80 E-2	248	-

("DF" is based on the calculation results)

Test of Continuous Treatment

Method of Test. The purpose of this method is to verify the removal performance of radioactive material and COD by treating test water with the membrane separation activated sludge process device. First, we prepared five kinds of test water as indicated Table IV, and added 0.1g/l of glucose as nutritious supplement for sludge into septic tank (sludge density: 15000mg/l). Then, we treated at 20l/d flow rate for 6 hours and passed it through the filtration membrane (0.45μm).

- Test water 1: Washing four uncontaminated uniforms with 20 l of water, adding 2g of detergent and also adding some crud to make 5-10Bq/ml radioactive concentration of test water.
- Test water 2: Washing four uncontaminated uniforms with 20 l of water, adding 2g of detergent and also adding slightly concentrated waste water to make 5-10Bq/ml radioactive concentration of test water.
- Test water 3: Washing five (low-level) contaminated uniforms with 20 l of water containing 2g of detergent and adding some crud to make 5-10Bq/ml radioactive concentration of test water.
- Test water 4: carrying out the pretreatment with 50mg/l of sodium hypochlorite for test water 2.
- Test drain 5: carrying out the pretreatment with 50mg/l of sodium hypochlorite for test water 3.
- T-Co: Total Co concentration
- S-Co: Soluble Co concentration (Co after filtration with 0.45μm membrane)
- Filtration DF: (T-Co)/(S-Co)

Table IV. State of Test Water

Test water No.	T-Co (Bq/ml)	S-Co (Bq/ml)	Filtration DF	COD (mg/l)	SS (ppm)	Electric conductivity (mS/cm)
Test water 1	8.40E+0	5.50E-2	153	13	20	0.15
Test water 2	9.44E+0	8.32E-2	113	27	174	1.70
Test water 3	7.02E+0	4.19E-2	168	58	32	0.30
Test water 4	8.66E+0	3.90E-2	222	27	106	2.20
Test water 5	1.06E+1	2.05E-2	517	109	67	0.63

("DF" is based on the calculation results)

Test Results. The results of the test are compiled in Table V.

- The results show that the radioactive concentration of all test water was reduced sufficiently (DF>100), and as for test water 1, 3 and 5, it was reduced remarkably (DF>1000). In addition, we determined that test water 2 and 4, which added concentrated wastewater, had sufficient DF performance from the results (DF>400, without

pretreatment by sodium hypochlorite; DF>700, with it; although these DF didn't increase for the other test water.)

- We determined that the effect of sodium hypochlorite increased the DF approximately twice by comparison between test water 2, 4 and 3, 5.
- We saw good performance for removal performance of COD in that all test waters were under 70ppm.
- It is clear that SS concentration of treated water is extremely low in consideration of the difference between filtration membrane for analysis (pore size: 0.45μm) and separation membrane in septic tank (pore size: 0.4μm). However, we checked the result of test water 3 just in case, and then recognized SS was less than 1ppm.

Table V. Results of Drain Treatment TEST

Test water No.	T-Co (Bq/ml)	S-Co (Bq/ml)	T-DF (-)	S-DF (-)	COD (mg/l)	SS (ppm)	Electric conductivity (mS/cm)
Test water 1	7.44E-3	7.44E-3	1129	7.4	5	-	0.25
Test water 2	2.08E-2	2.08E-2	454	4.0	9	-	1.30
Test water 3	5.12E-3	5.12E-3	1371	8.2	37	<1	0.40
Test water 4	1.12E-2	1.12E-2	773	3.5	25	-	1.90
Test water 5	4.21E-3	4.21E-3	2518	4.9	66	-	0.73

("DF" is based on the calculation results)
T-DF: Total DF(=(Before treatment T-Co)/(After treatment T-Co))
S-DF: Soluble DF (=(Before treatment S-Co)/(After treatment S-Co))

Technology of Laundry Drain Treatment (Ozone Oxidation Treatment)

Principle/Characteristics.

The purpose of ozone oxidation treatment is to oxidize organic material and produce carbon dioxide gas etc. by activated oxygen (such as OH radical etc.), which is generated by the decomposition of ozone injected in laundry drain liquid received in the ozone treatment tank. The reaction rate of the OH radical is 40 figures higher and better than O_2, and also 6 figures higher and better than O_3. As for the generation of this OH radical, it is possible to increase its treatment speed by extension of reaction area and selecting a catalyst, whose particle size is small, in order to set the fluidized bed because the natural decomposition speed of O_3 is too slow. A filtering device, using a ceramic filter, is installed for collection of catalyst and sludge because catalyst in the ozone treatment tank is discharged together with treated waste water outside of this tank. Sludge and spent catalyst, which are collected with the strainer, are recovered periodically and burned in an incinerator. An outline of the ozone oxidation treatment system is shown in Figure 6.

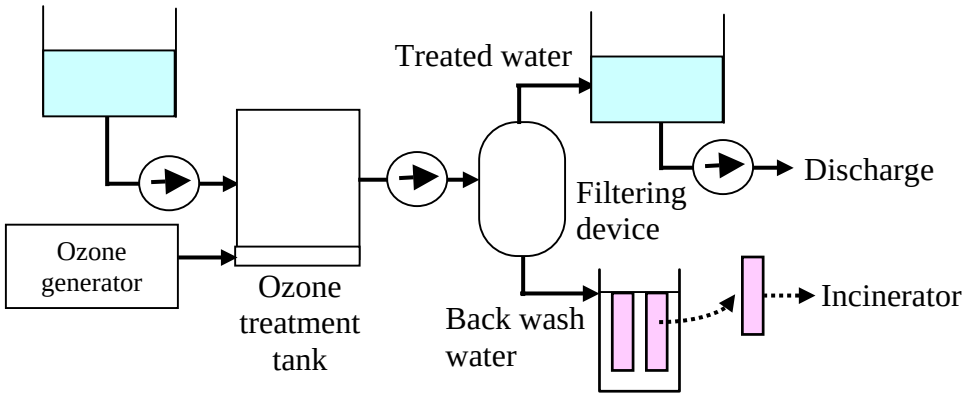


Fig. 6. Outline of the Ozone Treatment System

Hot Test

Method of Test. Four test waters were treated by ozone oxidation as indicated in Table VI. The parameters of radioactivity, COD etc. were controlled by adding slightly concentrated waste water or other components as necessary after washing contaminated or uncontaminated uniforms with water containing detergent similar to the activated sludge process tests. These test waters were treated at 80°C (reaction temperature) for 2 hours (reaction time) in the ozone treatment tank and filtrated with a 0.45μm millipore filter.

- Test water 1 and 2: Washing five (low-level) contaminated uniforms with 30 l of water containing 30g of detergent and rinsing with 160 l of water. Then separating these drains (laundry drain and rinsing one) into two containers equally, and adding water into each container. Finally, adding 5cc and 120cc of concentrated waste water into them, one is called test water 1 and another one is called test water 2.
- Test water 3: Washing five uniforms, which were dry-cleaned with water containing 2g of detergent, and adding some crud to make 3Bq/ml radioactive concentration of test water.
- Test water 4: Washing one uncontaminated uniform with 5 l of water and then adding 0.5g of detergent. Finally, adding some crud to make 6Bq/ml radioactive concentration of test water.

Table VI State of Test Water

Test water No.	Radioactive concentration (Bq/ml)	COD (ppm)	SS (ppm)	Electric conductivity (mS/cm)	Filtration rate (ml/sec)
Test water 1	6.37E-1	68.4	25	0.501	0.15
Test water 2	1.12E+1	74.8	138	1.004	0.18
Test water 3	3.38E+0	7.6	9	0.138	0.19
Test water 4	6.53E+0	18.4	32	0.147	7.00

Test Results. The results of the test are compiled in Table VII.

- This method gave excellent results, in that all test water achieved DF>100, in the reduction of radioactive concentration. It is thought that the DF of test water 3 didn't increase compared to other test water due to residual dry-cleaning detergent.
- As for COD removal performance, we got the good results that all test waters were below 30ppm.
- We got the excellent results that all test water achieved SS<4ppm.
- We determined that other test waters increased from 4 to 20 times, however, test water 4 was excluded from evaluation because its undiluted solution stood out from others.

Table VII. Results of Drain Treatment Test

Test water No.	Radioactive concentration (Bq/ml)	DF	COD (ppm)	SS (ppm)	Electric conductivity (mS/cm)	Filtration rate (ml/sec)
Test water 1	4.35E-4	1465	2.0	<4	0.446	0.57
Test water 2	7.16E-3	1561	22.9	<4	0.948	1.00
Test water 3	6.72E-3	503	0.1	<4	0.161	3.53
Test water 4	2.81E-3	2324	3.4	<4	0.165	6.67

("DF" is based on the calculation results)

CONCLUSION

Technology of Spent Resin Volume Reduction and Stabilization Process

We determined that the treatment of spent resin with this technology was affected by metal ions and crud. However, we verified that the performance (reduction of volume and weight of spent resin, conversion to inorganic material, detoxification, retention and transfer of nuclides) was sufficient to make it practicable.

Technology of Laundry Drain Treatment

We believe that these technologies of "membrane separation active sludge process" and "ozone treatment" can be put into actual use without any difficulty because we had excellent results with both technologies achieving the targets (DF>100, COD<90, SS<100).

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