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Development of the Pilot System for Radioactive Laundry Waste Treatment Using UV Photo - Oxidation Process and Reverse Osmosis Membrane

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Abstract

The pilot system for radioactive liquid laundry waste was developed with treatment capacity, 1ton/hr and set up in the Yong Kwang unit #4. The system is composed of tank module, RO systems and a UV/H_2O_2 photo-oxidation unit. The RO system consists of the BW unit (low-pressure RO for brackish water desalination) and the SW unit (high-pressure RO for seawater desalination). The BW unit possesses 4 RO membranes and it can reduce the feed water volume down to 1/10. This concentrated feed water can be reduced again up to 1/10 in its volume in the SW unit composed of 4 RO membranes. The UV/H_2O_2 photo-oxidation process unit was used for the detergent degradation. The operation of the pilot system was carried out and verified in its capability through the continuous operation and concentration operation using the actual liquid waste from the power plant. The design criteria and data for industrialization were yielded. The efficiency of the UV/H_2O_2 photo-oxidation process and the optimum operational procedure were evaluated. The decontamination factors for radioactive cobalt and cesium were measured. This on-site test showed the experimental result in the DF$\geq$ 300 and volume reduction factor$\geq$100.

Key Words : radioactive laundry waste, UV photo-oxidation, reverse osmosis, DF, detergent removal, radioactive Co, Cs separation

1. Introduction

Since the nuclear power plants are getting old and are increasing in number, the generation of radioactive liquid wastes is getting more in volume and various in type. Radioactive liquid laundry waste generated from the shower and laundry of the man working in the restricted area contains relatively low radioactivity ranged from 10^{-7} to 10^{-4} μCi/cc. However the yearly generation volume becomes up to 4000 ton per each reactor. This volume covers a large portion of the whole liquid radwaste released to the environment. Thus despite of its low activity, it contributes large part of the total radioactivity released to the environment from the nuclear power plant[1]. Accordingly, the radioactivity of laundry radwaste can not be disregarded in respect of environmental protection. Together with it, to achieve the goal of the "Zero-Release of the radioactivity from liquid waste stream, which is schemed by KEPCO to keep clean environment, the effective and safe liquid laundry waste treatment technology development is thus highly required[2].

The current radioactive liquid waste treatment process depends mainly on the evaporation system. The liquid waste is evaporated and concentrated, and the concentrate can be stored in a storage after solidification with cement or immobilization with paraffin. However radioactive liquid laundry waste is not desirable to be processed in the evaporator because of not easily degradable detergent in the waste water which can cause problems during the operation of evaporator and of its large volume. Accordingly, laundry radwaste water is presently being released to the environment after dilution with radioactivity below restricted level. However, to reduce the total radioactivity released to the environment, and to solve the above noted problems with evaporator, the development of the liquid laundry radwaste treatment process is required. The present study is, therefore, focused on the development of the process using reverse osmosis membranes which can separate radionuclides and detergent from the radwaste water without phase change.

The generalized on-site test was carried out in Yong Kwang # 4 with the pilot plant as shown in figure 1 since it was designed after the previous experimental results.

2. Material and Method

Both simulants and actual radwaste were used for the experiments instead of using only the actual laundry radwaste. Because the use of detergent is strictly limited in the nuclear power plant in order to prevent potential problems caused by detergent. For this reason, no foam was formed in the laundry waste from Yong Kwang #4. which was hard to observe the degradation effect of detergent in the UV reactor.

2.1. Material

A waste water with detergent was simulated with PF(commercial non-ionic detergent, 14%, supplementary agents 86%), and radionuclides in the tab water. The non-ionic detergent, consists of Lauryl Alcohol-9, 13.9%, Alcohol Ethoxylate, 1.5% The hydrogen peroxide, 30wt% was used as an oxidizer. For the test with actual radwaste, the laundry radwaste in Yong Kwang #4, which was kept in chemical waste tank, was used.

2.2. Method and Procedure

2.2.1. Test with Simulants

As shown in Fig. 1, the liquid waste in BW(Brackish Water) tank was processed with UV reactor and RO membrane through the two different flow paths. One flow path was connected in circulation mode from the BW tank to the UV reactor, or vice versa. The other one was from the BW tank to BW RO module. The waste water in BW tank flow passed through the micro filter and to the BW module after bypassed the activated carbon bed and carbon filter.

The test was carried out with simulants composed of tab water, 800 l , and detergent, 400g, of which TOC concentration was equivalent to 100 ppm. The simulants were filled in BW waste tank. Radionuclides, cobalt and cesium, were added into the simulants to be $10^{-5}\mu$Ci/cc in

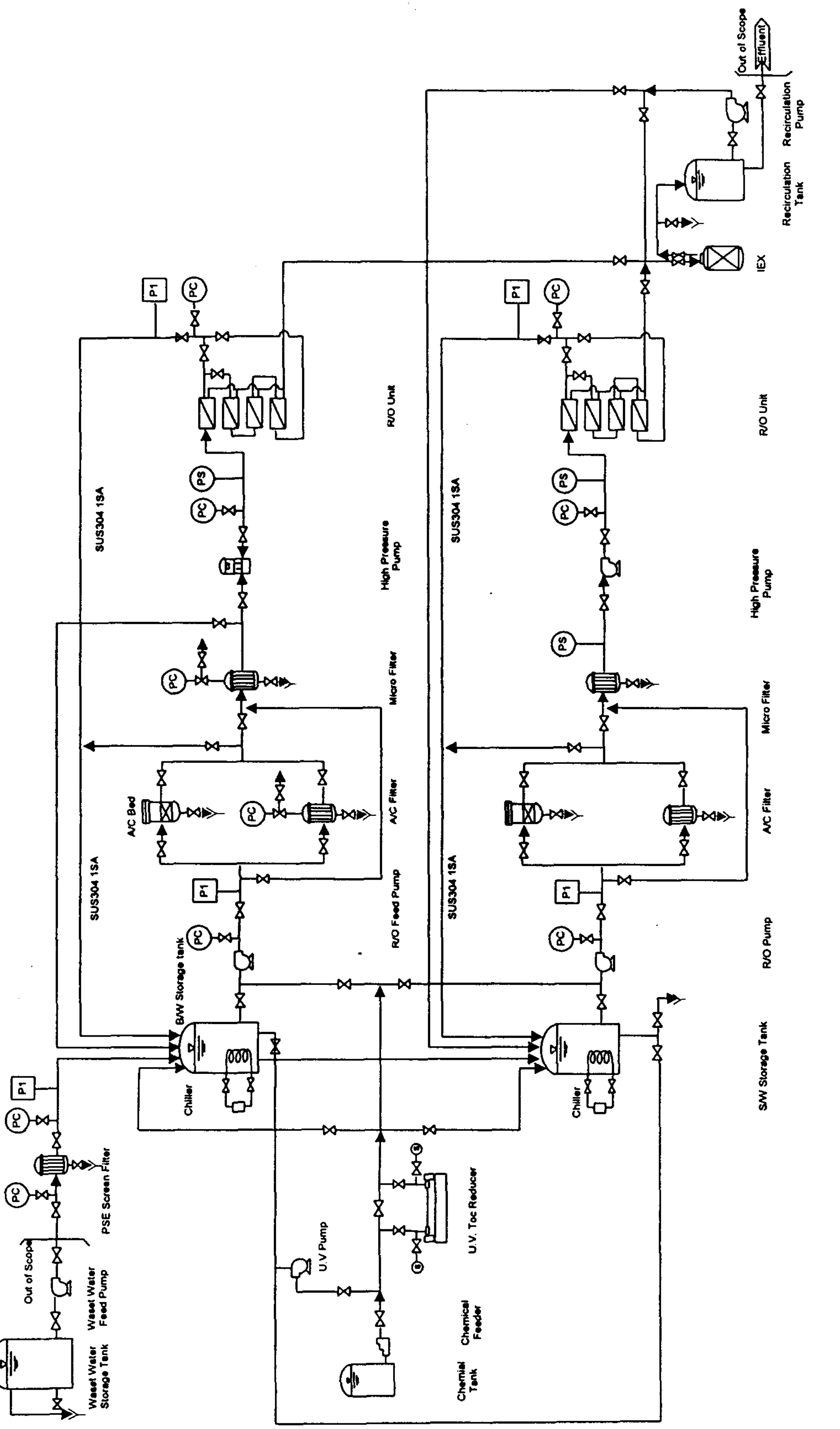

Fig. 1. P & ID of the Pilot Plant

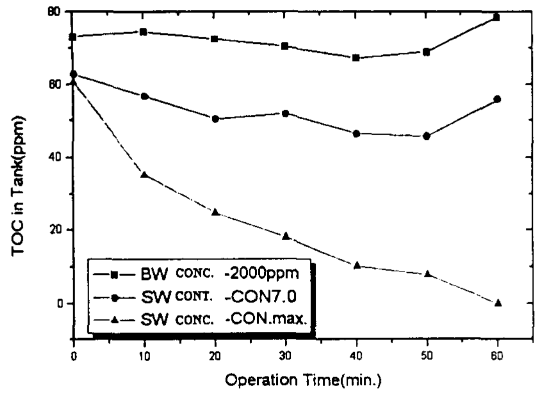

Fig. 2. The Result of Demonstration Test with Simulants

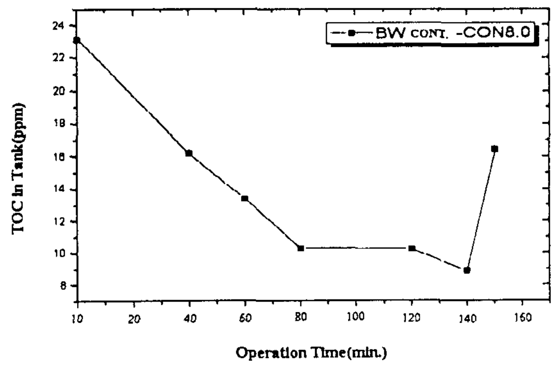

Fig. 3. TOC Variation of Actual Waste in the BW Tank during the Demonstration Test

activity. Hydrogen peroxide of 2000 ppm was added in the BW tank in a bulk mode. The concentration operation in the BW module was carried out with the permeate flow rate of 10 ~ 12 LPM and the feeding water pressure of 15 kg/cm^2. The operation was continued until the 800 l of waste water was reduced to 200 l . One liter of permeate was sampled from time to time for measurement of decontamination factor. The concentrated 200 l was sent to the SW RO module for volume reduction since feeding valve was closed. The operation under the closed condition was continued until the water was reduced to 50 l . For this test, hydrogen peroxide was added continuously under the adjustment in maximum or at 7.0 of the pump control nod of chemical injection kit.

2.2.2. Test with Actual Radwaste

All the lines were connected as the same as the simulants test except the line-up for continuous feeding to BW tank from chemical waste tank. First of all the actual laundry radwaste, 800 l from the chemical waste tank, was filled in the BW module tank. The feeding valve was closed since the feeding water was filled in the tank. Radionuclides, Co-60 and Cs-137, were added as a tracer, into the tank in radioactivity, 10^{-5} μCi/cc respectively. And then the UV reactor and BW membrane module were operated. At the same time the feeding valve was opened, and the flow rate of the feeding water was controlled at 10 LPM which was the same as that of the permeate.

3. Results

3.1. Demonstration Test with Simulants

The 800 l of simulants with the initial TOC 73ppm was filled in the BW module tank for concentration operation. During the concentration of the radwaste water to 200 l , the samples for TOC measurement were taken every 10 minutes. Slight increase in the TOC from 73 to 78ppm was observed when the concentration operation was finished. Bearing in mind, it seems that the large portion of TOC was removed in the UV reactor, otherwise the final TOC after concentration should be about 290ppm.

The line with closed square in Figure 2 indicates the variation in TOC of the waste in the BW tank during the concentration operation. This figure shows that the TOC decreases for 40 minutes

Table 1. Radioactivity and Decontamination Factor in Concentrate and Permeate

Radioactive Co			Radioactive Cs		
Concentrate	Permeate	DF	Concentrate	Permeate	DF
1.46×10^{-5}	4.98×10^{-8}	292	5.23×10^{-5}	1.67×10^{-7}	313
1.01×10^{-5}	3.57×10^{-8}	282	4.11×10^{-5}	1.36×10^{-7}	302

since operation started, and then it increases. This fact indicates that hydrogen peroxide injected in a pulse mode could be almost exhausted within 40 minutes and also indicates that the UV reactor was effective in reducing TOC.

The operation of SW RO module was performed after the 200 l of concentrates was discharged from BW tank to SW tank. The line with closed circle in Figure 2 indicates TOC variation of the waste in the SW tank. Hydrogen peroxide was continuously added during the operation. While the operation was continued for an hour, TOC was decreased but evenly. This is the evidence for that the UV reactor could remove TOC under the above condition. Besides, the line with closed triangle indicates TOC variation during the concentration in the SW module under the closed condition which means the feeding valve was closed. The TOC was decreased in linear pattern. This means that TOC can be nearly zero at the end of the operation.

3.2. Demonstration Test with Actual Radwaste

Figure 3 shows the TOC variation of the actual laundry waste which was concentrated for 50 minutes in BW module. Before concentrated, 1.2ton of waste water was treated in BW module under the continuous operation for 120 minutes with the flow rate, 10 LPM for both feeding and permeating. Contradictorily, the initial TOC was 23ppm when the concentration began, despite the initial TOC of radwaste preserved in the chemical waste tank was 5.5ppm. This difference could be occurred by the high concentration of residual TOC inside the pipes. The TOC in the figure was reducing in linear pattern. This tendency indicates that the UV reactor worked effectively for the TOC removal.

During the BW RO operation, the fouling of the micro-filter was occurred frequently. This fact interrupted the smooth operation. Therefore, the micro-filter which can backwash during the operation is required to prevent the operational interruption.

3.3. Decontamination Factors for Radioactive Cobalt and Cesium

Despite of the radioactivity of concentrate, 10^{-5} μCi/cc, the activity of permeate was lower than LLD (Lower Limit of Detection). The DF was about 300. Table 1 shows the radioactivities and DFs of cobalt and cesium.

4. Conclusions

The 5μm pore filter which was used in the process was fouled frequently, therefore, it is concluded that the present filtration system has to be changed to the backwash filtration system for the smooth operation of RO process. The volume reduction factor over 100 in the RO module was achievable through this test. The DF for Co and Cs were about 300 each. The radioactivity of radwaste water was about $10^{-5}\mu Ci/cc$, that of the permeate was lower than LLD.

The followings are suggested as the design criteria for industrialization.

- Design criteria included the capacity of 1ton/h, the DF≥300 and volume reduction factor≥ 100,
- Pretreatment filtration is desirable in composition with 25㎛ and 5㎛ pore sized filters.
- Two of each 25㎛ and 5㎛ filters can be arranged in a row, otherwise one of each backwash filter is recommendable.
- The backwash type using air pressure is better in respect of reducing secondary waste, and the material of it is desirable in the inorganic material such as stainless steel.

The UV/H_2O_2 photo-oxidation reactor is better designed for TOC removal more than 70% of every waste water passed one time through. The wave length range of UV lamp is desirable in between 254nm and 400nm[7]. The automatic cleaning wiper on quartz sheath of UV lamp is necessary. H_2O_2 injection kit with a pump of broad injection volume (0-5 LPM) and continuous type (not pulse type) are also required.

For the RO membrane module, using only the eight of low-pressured BW module, spiral-wound type made of polyamid gives more economical benefit than using together with SW module. Piping has to be concerned of separation of the flow path between the UV reactor and the RO module. Operational point of view, water level has to be kept constantly until one laundry waste water tank being empty. The concentration operation is better to be started after the previous water tank empty. Hydrogen Peroxide has to be controlled in higher concentration for concentration operation than that for continuous operation. The flow rate is better keeping higher to UV reactor than to RO module.

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