Treatment process of waste water from UF6 cylinder washing process in a low temperature and low pressure

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I. Introduction

UF_6(Uranium Hexafluoride, less than 5w/o of low concentrate uranium) used as nuclear fuel in a pressurized water reactor is generally contained and transported in a 30B Type of special cylinder, because it can react with moisture in atmosphere, produce hazardous uranyl fluoride(UO_2F_2), and hydrogen fluoride(HF).

To reuse the cylinder, therefore, it should be washed and inspected under the domestic nuclear law and international transportation regulations (1, 2). Chemical wash, physical wash and electrolytic wash are used for washing the cylinder. The most widely used method is chemical wash.

The cylinder wash process studied in this paper washes cylinder in chemical method with demineralized water, Na_2CO_3 and H_2O_2. In the process, 200L/batch (20 m^3/yr) of liquid radioactive waste is produced and the waste contains $Na_4UO_2(CO_3)_3$, UO_2F_2, $Na_2U_2O_7$ and NaF etc. (1, 2, 3)

The liquid waste is treated in this procedure; 1) Boiling the liquid waste at 70℃ for removal of carbonate. 2) Add NaOH in the wastewater and filter the mixture with a screen. 3) Screened residue is dried and then stored in a uranium storage. 4) liquid part is moved to a storage tank and radioactivity is measured in the liquid. 5) If the concentration of radioactivity is lower than corresponding regulation limit, the liquid moved to a reaction tank and evaporated with additional low concentration HF in 105℃. 6) Radioactivity of distillate is measured and the value is lower than regulation, it is treated with a thermal decomposition process and discharged to the atmosphere in gas state. 7) Solid waste produced in the evaporation step is managed as solid nuclear waste.

The treatment procedure has several shortcomings as high operating expenditure because of boiling at 105℃, and producing large amount of solid waste by using of NaOH and low concentration's HF.

In this study, it is investigated that the liquid waste from the real scaled cylinder wash process can be treated by low temperature and low vacuum of evaporation system to confirm the feasibility of application under the emission standard, and reduction of waste production as well as energy consumption.

I.A. Test method

I.A.1. Evaporation test

The evaporation test was conducted using a lab-scaled evaporation equipment composed of Pressure Regulation System (SC-820), Temperature Control System (DMSD-635) and Coolant Circulator (CCA-111).

Tests were conducted at low pressure(30mbar) and low temperature(35℃). By rotating with a magnetic stir bar (250RPM), the sample in the equipment was heated uniformly. The evaporation test was conducted in triplicate.

I.A.2 Gross alpha analysis

Radioactivity concentration was determined by a gross alpha analyzer equipped with proportional counter detector (FHT 8000A measuring channel/Thermo). Planchet used in the analysis was 60mm and 10mL in diameter and volume, respectively.

I.A.3. *Gamma Radionuclide Analysis*

Gamma radionuclide in the sample was analyzed by HPGe (High-purity Germanium detector, CCII-VD, CANBERRA) with cylindrical beakers (20/80mL).

Fig.1. lab-scaled evaporation equipment

I.A.4. *XRF Analysis*

Elemental composition of sludge was analyzed by XRF(X-Ray Flourescence Spectrometry, S8 Tiger, Bruker) with sample cup of mylar and polypropylene.

I.B. Test result

I.B.1. Radioactivity removal efficiency

The radioactive concentration removal in gross alpha test was 99.9% in all batches and it is possible to reduce the concentration in the distillate below the discharge regulation (0.08Bq/mL). (Table Ⅰ.)

TABLE I. Radioactive concentration and pH in raw liquid waste and distillate.

Items		Raw liquid waste	Distillate	Note
Gross alpha analysis (Bq/mL)	1	116.3	N/D	
	2	131.5	N/D	
	3	71.0	N/D	
Gamma Radiation (Bq/mL)	1	31.5	N/D	U-235
	2	43.6	N/D	
	3	34.8	N/D	
pH	1	8	7	
	2	9	7	
	3	8	7	

1. Supernant

16 kinds of radionuclides including U-235, Pa-234M, Th-231, I-126 and BI-211 were detected in the wastewater. In the distillate, however, the radionuclides of uranium was detected under the MDA(about 0.002Bq/mL) and the gamma radiation was not detected, respectively. The removal efficiency of radionuclides is over than 99.9%. (Table 1.)

The glass fiber was installed for solving the problem as residual radioactivity in distillate. Because the radionuclides during evaporation were included in the foam and could be attached in the fast vaporization compounds as CO_3 and CO_2. After filter installation, the concentration of gross alpha as well as gamma in distillate were detected as below MDA, respectively. (Table 1.)

I.B.2. Secondary waste

The chemical treatment process had produced average 15drum/yr of secondary waste by additional chemicals as 2ton/yr of NaOH and 10ton/yr of HF. On the other hand, the evaporation system is not necessary the chemical addition and has produced 15g/L of sludge. Therefore, it could be expected the 1.5drum/yr of secondary waste production and about 90%(13.5ton/yr) waste reduction. The final waste sludge will be stocked in the special uranium storage area. Henceforward, it would be made a waste or self-disposal via uranium extract process. In addition, the elemental components in final sludge are mainly U and Na by XRF Analysis. (Table Ⅱ.)

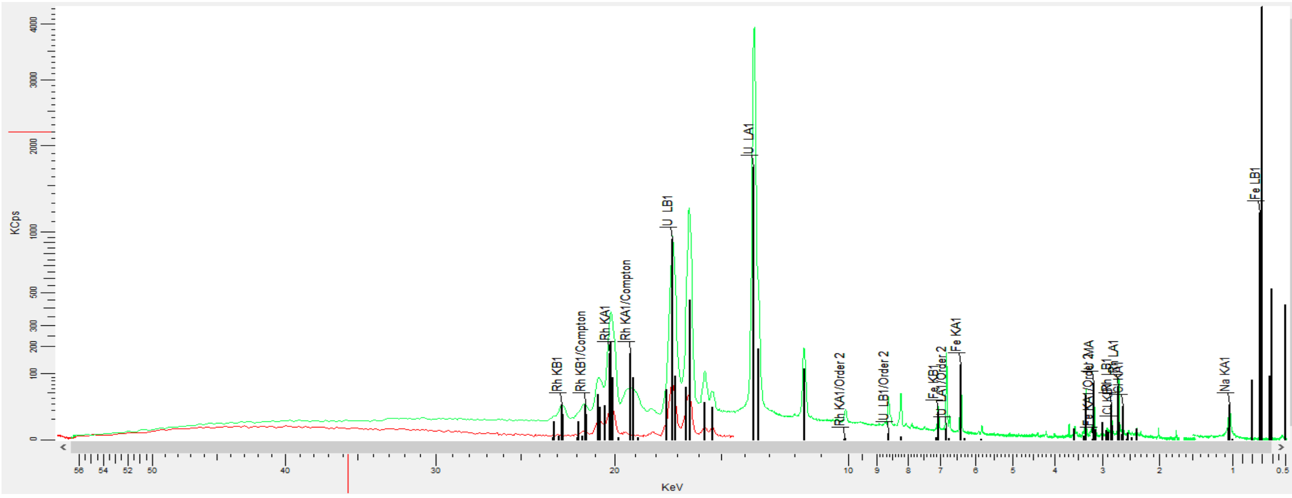

Figure 2. Result of XRF Spectrum

TABLE 2. Elemental composition of final sludge.

	Element	Conc(%)
1	U	45.5
2	Na	43.0
3	Fe	3.23
4	Rh	3.17
5	Cl	2.13

I.B.3. pH

The pH of distillate after evaporation was neutralized. Even though, the pH of raw wastewater was between 8 and 9 (Table 1.)

II. CONCLUSIONS

The treated water by low temperature and low pressure evaporation system from UF6 cylinder wash process had very low concentration of gross alpha under MDA and could be treated under 0.08Bq/mL of allowable uranium discharge standard. Not only the radionuclide in gamma radiation were removed over 99.9% by the evaporation system, but also the 16 kinds radionuclide were removed completely.

And the secondary waste production from evaporation system was 10% of exist wastewater treatment process. Thus it is expected to reduce the waste production 90%(from 15drum/yr to 1.5drum/yr) by converting treatment system form the chemical method to evaporation system.

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