Washing water treatment process for UF₆ cylinder by adjusting evaporation technology in a low temperature and low pressure

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1. Introduction

 UF_6 (Uranium Hexafluoride) used as nuclear fuel in a light-water reactor is generally contained and transported in a special cylinder, because it can react with moisture in atmosphere and 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, Na₂CO₃ and H₂O₂. In the process, 200L/batch (20 m³/yr) of liquid radioactive waste is produced and the waste contains Na₄UO₂(CO₃)₃, UO₂F₂ and NaF etc. (1, 2, 3)

The liquid waste is treated in this procedure; 1) Add NaOH to the liquid waste and filter the mixture with a screen. 2) Screened residue is dried and then stored in a uranium storage. 3) liquid part is moved to a storage tank and radioactivity is measured in the liquid. 5) If the of radioactivity concentration is lower than corresponding regulation limit, the liquid moved to a reaction tank and evaporated with additional low concentration HF in 105 $^{\circ}$ C. 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 mentioned above has disadvantageous points, producing large amount of solid waste as well as, high energy and chemical consumption.

In this study, liquid waste from a real scaled cylinder wash process is applied to evaporation system to confirm feasibility of the application of evaporation and, to reduce waste production and energy consumption.

2. Test method

2.1 Evaporation test

The evaporation test was conducted using an labscaled 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(40 $^{\circ}$ C). By rotating with a magnetic stir bar (250RPM), the sample in the equipment was heated uniformly.

The evaporation test was conducted in triplicate.

2.2 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.

2.3 Gamma Radionuclide Analysis

Gamma radionuclide was analyzed by HPGe (Highpurity Germanium detector, CCII-VD, CANBERRA) with marinelli beakers (450mL).



Fig. 1. lab-scaled evaporation equipment

3. Test result

3.1 Radioactivity removal

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.)

Items	Batch number	Raw liquid waste	Distillate
Gross alpha analysis (Bq/mL)	1	71.8	0.08
	2	71.8	0.03
	3	19.2	0.01
U-235 (Bq/mL)	1	43.8	7.9E-03
	2	43.8	8.6E-04
	3	13.9	1.2E-03
рН	1	11	8
	2	11	7
	3	10	7

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

The first batch has higher gross alpha value, 0.08Bq/mL, than other batches. This is because some concentrate flowed in the distillate with brisk reaction in the evaporation vessel.

Thus, the second and third batches were conducted with a glass fiber filter in the top of the evaporation vessel. The radioactive concentration of distillate from the second and third batches were 0.03 and 0.01Bq/mL, respectively. (Table 1.)

16 kinds of radionuclides including U-235, Pa-234M, Th-231, I-126 and BI-211 were determined in the liquid wastewater. In the distillate, however, only trace amount of U-235 was detected. The removal efficiency of radionuclides is over than 99.9%. (Table 1.)

3.2 Secondary waste

The chemical treatment process produces 15drum/yr of secondary waste with 2ton/yr of NaOH and 10ton/yr of HF addition. On the other hand, evaporation treatment system does not need additives and produced 15g/L of sludge. Therefore, it is expected that 1.5drum/yr of secondary waste production and 90% waste reduction.

3.3 pH

With the evaporation treatment, pH of the water was neutralized. pH of raw wastewater were between 10 and 11, but pH of distillate were 7 and 8 (Table 1.)

4. Conclusions

Liquid radioactive wastewater from a real scaled UF6 cylinder wash process was applied to evaporation treatment system.

Radioactive concentration in gross alpha was removed 99.9% in the evaporation system. And the concentration in distillate was lower than the discharge regulation.

Removal of U-235 was 99.9% in the process. And 15 other kinds of radionuclides in the raw wastewater were removed completely.

Secondary waste production of the evaporation system is 15g/L. Thus it is expected to reduce waste production 90% (from 15drum/yr to 1.5drum/yr) by converting the treatment system form the chemical method to evaporation system.

Reference

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