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The development and construction of the commercial uranium purification plant

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1. Introduction

In the nuclear fuel cycle, uranium purification is the most important technology applied across the board; refining, conversion, fuel fabrication, and reprocessing. Besides, it can be applied for decontamination and decommissioning, in terms of the recovery of uranium from wastes [1].

As a part of the uranium refinement and separation technology development project, we, the engineers at KEPCO Nuclear Fuel (KNF) have been researching into a uranium purification process and have designed the process through LAB test independently.

As a result of this research, we have developed several process equipment and control method, and constructed the commercial uranium purification plant in the KNF field by our own EPC (Engineering, Procurement, and Construction) work utilizing these results. And we have performed the test operation to verify the performance of this plant, and met encouraging results. This process is described herein, and the test operation procedure and representative results are presented below.

2. Process description

In this section the uranium purification process developed and constructed by KNF is described.

2.1 Overall features

As a purification method, the solvent extraction has been chosen as other companies use. The uranium purification process consists of two unit processes; solvent extraction process and solvent regeneration process [2], and only the former one is addressed in this article.

Uranium (Ⅵ) is fed and discharged in the form of uranyl nitrate hexahydrate (UNH) aqueous solution, and the extractant, organic solvent is 30 vol% tri-N-butyl phosphate (TBP) in dodecane. The maximum capacity of this plant is 5 kg·U/hr.

The solvent extraction process includes three pulsed columns - one extractor, one scrubber, and one stripper, and several buffer tanks for discharges of each phase from the pulsed columns, transfer pumps - especially metering pumps for injection of each phase to the pulsed columns. The patented unique technologies of the developed process are the construction of the pulsed columns, the pulse generator, and the interface control unit.

2.2 Pulsed columns and pulse generators

As described, the uranium purification plant of KNF has three pulsed columns. All columns are rectangular with 225 ㎠ inner cross sectional area. Each column consists of top head, column body, and bottom head. A column body has inner structures assembled with a number of perforated plates with 1 ㎜ thickness. The perforated plates are designed using dispersed drop size estimation models to yield good interfacial area.

Each pulsed column has one pulse generator, a diaphragm pump connected to the bottom head [3]. The frequency of each pulse generator is controlled by an inverter, the pulse amplitude by a dial knob on each pulse generator.

2.3 Interface control

The most important technology in operating this process is the control of the interface between the organic solvent (organic phase) and the aqueous solution (aqueous phase) in the top head of a column. The process of KNF has the interface control device for each pulsed column as shown in Fig.1, and consists of three parts; level switches, interfacial level transmitter, and interface control units. The column has several level switches, one interfacial level transmitter, and one interface control unit.

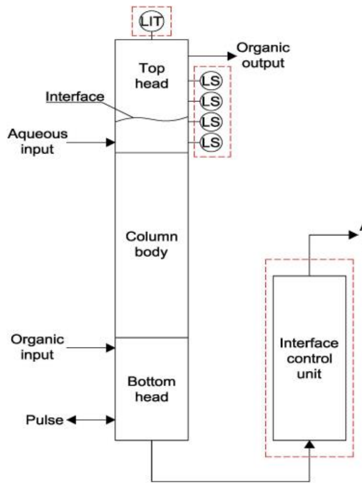

Fig. 1. Scheme of the interface control device

There are four level switches on the top head, placed along the wall sides of the heads. These level switches show the level range the interface is formed.

The interfacial level transmitter is placed on top of the top head. This level transmitter directly detects and displays the level of the interface.

The interface control unit maintains the level of the interface in a certain range using the pressure difference between the column and the unit itself [4].

3. Test operation procedure

To verify the uranium purification performance of this plant, we have performed the test operation. In this section the test operation procedure of the process is described.

3.1 Preparation

Chemicals to be fed to this process were prepared as shown in the Table I.

Table I: Preparation of chemicals

Chemical	Amount (litres)
Crude UNH solution	616
Fresh solvent	300
Nitric acid (3N)	240†

†Initial solution of pulsed columns was set aside

The crude UNH solution was prepared by dissolving uranium oxide powder containing some impurities in concentrated nitric acid. The fresh organic solvent was made of 30 vol% TBP in dodecane. All nitric acid (3N) used in this process was prepared by diluting concentrated nitric acid in demi-water.

Samples of each chemical were analyzed. The analysis items were as follows: impurity content for all, uranium content for the crude UNH solution, and the concentration for the concentrated nitric acid. The impurity content analysis results are presented in the Table Ⅱ, for the elements which levels in the crude UNH solution exceed the UO_2 powder specification limits.

Table Ⅱ: Major impurities in each chemical

Element	Crude UNH (ppm)†	Fresh solvent (ppb)	Nitric acid (ppb)	Demi-water (ppb)
Al	104.8	33	1,700	60
B	2.4	-	1,200	979
Ca	104.8	-	3,300	503
Cr	30.1	345	37,000	1
Fe	515.3	6	153,000	22
Mo	16.1	-	1,500	2
V	1.0	-	124	-
Zn	3.8	-	69	19.2
Total‡	4,500.5	9,370	213,515	3,910

†Average values of solution basis analysis results
‡Including other impurities

The uranium content of the crude UNH solution was 15.7 wt% (197.9 g·U/ℓ), and the concentration of the nitric acid was 68.5 wt%.

3.2 Process operation

3N nitric acid was filled in each pulsed column as initial solution, to the level of the lowest level switch on the top head. And the pulse generators were turned on before feeding each phase to the pulsed columns.

3.2.1 Purification of the crude UNH solution

There are three steps in the uranium purification process producing pure UNH solution satisfying the impurity specification of nuclear grade UO_2 powder, as shown in Fig.2; extraction, scrubbing, and stripping.

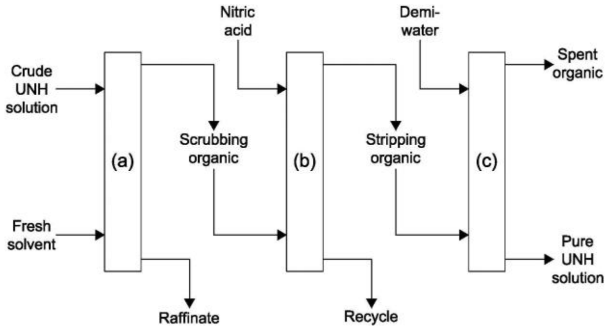

Fig. 2. Scheme of the interface control device; (a) Extractor, (b) Scrubber, (c) Stripper.

In the extractor, the first pulsed column, the direction of mass transfer of uranium (Ⅵ) was from the aqueous phase (crude UNH solution) to the organic phase (fresh solvent). The uranium-loaded solvent (scrubbing organic) overflowed from the top head to the scrubbing organic buffer tank.

In the scrubber, the second one, the direction of mass transfer of impurities was from the organic phase (scrubbing organic) to the aqueous phase (3N nitric acid) [5]. And the small portion of uranium (Ⅵ) in the organic phase was also transferred to the aqueous phase. The discharged aqueous phase was separately stored to be recycled to the extractor. And the scrubbed organic phase (stripping organic) overflowed from the top head to the stripping organic buffer tank.

In the stripper, the last one, the direction of mass transfer of uranium (Ⅵ) was from the organic phase (stripping organic) to the aqueous phase (demi-water). Both feeds were heated by double-pipe type heat exchangers for each feed before injection to the pulsed column. The organic phase lost uranium (Ⅵ) and discharged from the stripper (spent organic) was fed to the solvent regeneration process.

Pure UNH solution was discharged from the stripper, and was stored in the pure UNH storage tanks after cooled naturally in the buffer tanks. The pure UNH solution was sampled and analyzed. The impurity content analysis result showed that the levels of most elements were under the respective detection limits.

3.2.2 Test powder production and analysis

After all prepared crude UNH solution were purified, the test powder (UO_2) was produced from the pure UNH solution through AUH (Ammonium Uranate Hydrate) process. AUH process is the wet conversion process developed by us, KNF engineers, and includes main unit processes as follows; AUH crystallization, Liquid/Solid separation, and fluosolids reduction.

The produced test powder was sampled and analyzed in terms of uranium content and impurity content.

4. Results and discussion

The result of the test operation shows that the uranium purification plant of KNF is applicable to nuclear fuel manufacturing as described below.

Some parts of the impurity content analysis results are presented in the Table Ⅲ; impurities in the crude UNH solution, the test powder (UO_2) from the pure UNH solution produced by this uranium purification process, and the virgin powder produced by the commercial DC plant from UF_6, comparing with the UO_2 powder specification. The impurity levels and the equivalent boron concentration (EBC) in the test powder from the pure UNH solution satisfy the specification limits for all elements. In comparison to the virgin powder, the levels of four elements (Ca, Cu, Fe, and Zn) in the test powder are higher than those of the virgin powder and one element (Mg) is lower while others are not detected. The EBC of the virgin powder is 18 times of that of test powder.

Table Ⅲ: Comparison of major impurity content

Element	Spec. limits (ppm)	Crude UNH (ppm)†	Test powder (ppm)	Virgin powder (ppm) ‡
Al	50.0	588.4	-	0.8
B	1.0	13.4	-	0.13
Ca	50.0	588.4	16.46	4.48
Cr	50.0	168.8	-	1.22
Cu	50.0	27.1	0.21	0.1
Fe	75.0	2,893.2	10.21	5.3
Mo	20.0	90.3	-	1.08
V	1.0	5.8	-	0.1
Zn	20.0	21.4	3.62	0.1
EBC	2.36	15.89	0.01	0.18

†Converted values of solution basis into UO_2 basis
‡Average value of randomly selected 5 lots

For the crude UNH solution, the contents of 8 items exceed the specification limits; Al, B, Ca, Cr, Fe Mo, V, and Zn, and the average excess rate of these items is 1,028.4%.

For the test powder, the ratio of the impurity content to the specification limit for every element is lower than 40%, the highest one is Ca (32.9%), the lowest one among the detected impurities is Cu (0.4%), and the average value is 3.2%. The average rejection rate is 92.4%.

For the virgin powder, the ratio of the impurity content to the specification limit for every element is lower than 50%, the highest one is Bi (45.0%), the lowest one is Cu (0.2%), and the average value is 6.9%.

It is presented that most of the impurity levels and the EBC in the test powder are lower than those of the virgin powder. This is a very impressive result because the origin of the test powder is uranium scraps polluted by foreign materials from various sources while the DC virgin powder has no pollution source.

5. Conclusions

As described, we, the engineers at KNF have independently developed the uranium purification process and its devices; unique equipment and control methods which made the process simpler than the typical one. With this development results, we have constructed the commercial plant by our own EPC work, and performed the test operation. The results of the test operation show that the impurity levels of the most items in the test powder produced by the developed process are lower than that of commercial DC virgin powder made from UF_6. It denotes that the uranium purification plant of KNF is applicable to nuclear fuel manufacturing.

From now on, KNF business area can be extended to front-ends and back-ends of the nuclear fuel cycle by using this developed uranium purification process.

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