Understanding the PUREX Process with the Hanford Facility

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1. Introduction

For the denuclearization of the Korean Peninsula, an understanding of North Korea's sensitive nuclear cycle facilities must be preceded. Countries that operate nuclear facilities have their nuclear fuel cycles, and there may be differences in the scope and detailed processes of facilities that can be operated depending on the country's circumstances. However, in the case of North Korea, mining, refining, conversion, enrichment, processing, It has all nuclear fuel cycles of reprocessing. Among them, the nuclear cycle facility directly related to the conversion of nuclear weapons is an enrichment and reprocessing facility, which is essential for evaluating the nuclear transparency of the country. However, since it is difficult for a non-nuclear country to operate a sensitive nuclear cycle facility under the international nuclear non-proliferation regime, it is intended to simulate this through a virtual model and provide a framework of perception for evaluating the country's nuclear transparency.

2. Basic Theory

While Pu+3 has good extractability for the aqueous phase, Pu+4 has good extractability for the organic phase. Through this, the wet reprocessing process is based on purifying pure Pu through the oxidation-reduction of Pu ions. In particular, the method of dissolving Pu+3 with nitric acid and Pu+4 with an organic solvent (TBP, Tri-Butyl Phosphate) for purification is called PUREX. Since the target nuclear material of this study is limited to Pu, only the related processes will be presented briefly. The following table briefly presents the reaction equations used to calculate the mass balance in the main processes. [1], [2]

Table 1. Main process and reaction equations

Process	Chemical Equation
Dissolution	$Pu + 8HNO_3 \rightleftharpoons Pu(NO_3)_4 + 4NO_2 + 4H_2O$
Oxidation	$PuO_2^{+2} + NO_2^- + 2H^+ = Pu^{+4} + NO_3^- + H_2O$ $Pu^{+3} + NO_2^- + 2H^+ = Pu^{+4} + NO + H_2O$
Extraction	$Pu^{+4} + 4NO_3^- + 2TBP \rightleftharpoons Pu(NO_3)_4 \cdot 2TBP$
Reduction	$Pu^{+4} + Fe(H_2NSO_3)_2 \rightleftharpoons Pu^{+3} + Fe(H_2NSO_3)_2^{+1}$
Purification	$Pu + 8HNO_3 \rightleftharpoons Pu(NO_3)_4 + 4NO_2 + 4H_2O$

3. Model development

North Korea, a threat to the international nuclear non-proliferation regime, has a reprocessing facility using the PUREX process. The differences between the Hanford facility, the reference facility for this study, and the reprocessing facility in North Korea are as follows. [3]

- There will be significant differences in processing capacity but no differences in the flow of the process.
- Although there is a difference in the type of irradiated nuclear fuel, the difference will occur only in the jacket removal process, and there will be no significant difference in the subsequent process.
- The Hanford facility has an additional process to remove the fission product for safer work, and the North Korean facility is introducing a tertiary purification process to increase the purity of Pu. These processes may slightly affect the recovery rate of the Fission product or Pu but do not cause a big difference.
- Although there is a difference in the devices (Pulsed Column, Mixer-Settler) used in the extraction process, there is a difference in the processing capacity and no difference in the principle of the process.

As a result, there is no significant difference in the reprocessing process itself, and in conclusion, almost similar results will be obtained in the production of Pu. The amount of waste generated within the process will also have a similar result.

The spent fuel reprocessing facility is a sensitive nuclear cycle facility, so access to information is limited. Therefore, we designate the Hanford facility, which has relatively detailed information about the facility, as a reference facility and build a model based on it. The reprocessing process of the Hanford facility is divided into five parts, and the purpose and characteristics of each part are as follows.

- Solution preparation: chemical stripping, dissolution of spent nuclear fuel
- Primary solvent extraction: Removal of fission products through extraction-stripping
- U/Pu separation: U-Pu co-extraction, Pu extraction, U extraction
- U purification: U purification recovered through extraction-stripping
- Pu purification: Purification of recovered Pu through extraction-stripping

The operation method of the reprocessing facility is a mixture of a batch process and a continuous process, so it is necessary to operate by time step as a whole. In order to simulate this, the flow of material is optimized for each time step and configured. To this end, two solution preparation lines were configured so that the subsequent purification process could be operated without a rest period. Based on the Hanford facility operation data, the flow of materials required or generated for the operation was configured. The material balance was reconstructed based on this. The model was constructed to estimate the input material's number of by-products and products.

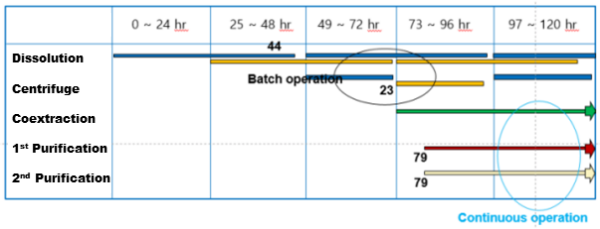

Figure 1. Schematic diagram of step analysis

4. Modeling Results

For detailed modeling of the process, a more straightforward and more intuitive P&ID (Piping and instrumentation diagram) was derived by extracting only the main equipment based on the P&ID information of the Hanford facility. Based on this P&ID information, the AnyLogic™ simulation model was developed. [4]

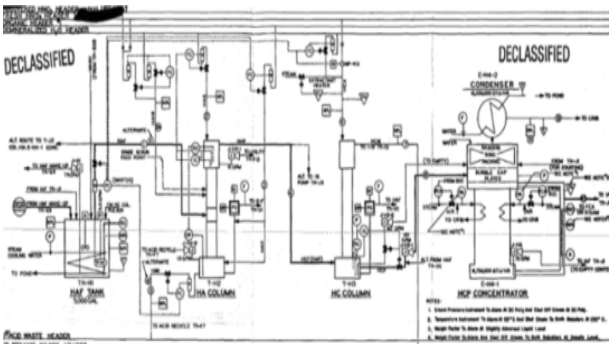

Figure 2. Real P&ID

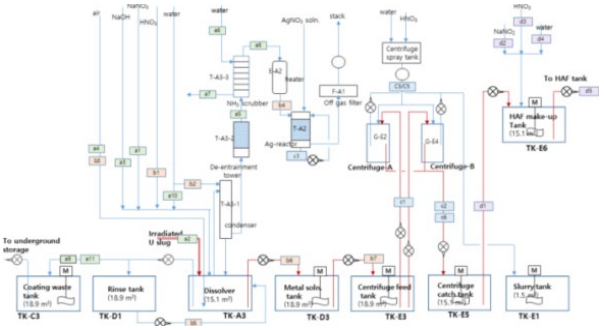

Figure 3. Simplified P&ID

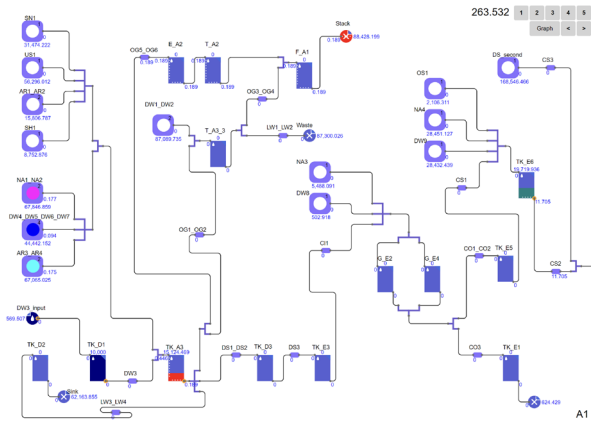

Figure 4. Process model using discrete event modeling (AnyLogic™)

Based on the material balance of the Hanford facility, a new material balance was derived by correcting the difference in overall capacity and some processes, and the model was configured to move sequentially according to the time step of the process. The following table is a part of the new material balance that includes a process similar to a radiochemical facility in North Korea.

Table 2. sample of material balance in process

unit process	flow (input)	calculated weight (kg)	calculated volume (liter)	from book weight (kg)	from book volume (liter)	flow (output)	calculated weight (kg)	calculated volume (liter)	from book weight (kg)	from book volume (liter)
dissolution	SN1	5245.70	4408.15	5246.25	4413.66	OG1	1016.22	G	N/A	
	US1	9382.67	S	9377.57		OG3	1030.70	G	1030.70	
	AR1	1013.26	G	1013.26		LW1	5568.20	5568.20	5597.85	5605.16
	OG1	1016.22	G	N/A		OG2	1699.49	G	N/A	
	DW1	5582.68	5582.68	5582.68	5582.68	LW2	8981.81	8981.81	8956.57	8968.25
	SH1	1458.81	1064.83	1458.75	1064.78	OG4	1649.96	G	1649.12	
	AR2	1621.21	G	N/A		LW3	6932.39	5332.61	7176.28	5545.46
	OG2	1699.49	G	1649.12		LW4	3428.25	3428.25	3530.31	3535.47
	DW2	8932.28	8932.28	8932.28	8932.28					
	DW3	3428.25	3428.25	3428.25	3433.27					
	NA1	6368.68	4648.67	6360.95	4655.92	OG5	6790.84	G	6795.27	
	DW4	3390.38	3390.00	3388.34	3393.52	DS1	14632.66	8507.36	14631.53	8520.71
	AR3	6295.29	G	6300.85		OG6	6790.84	G	6795.27	
	DW5	832.38	832.38	N/A		DS2	14632.66	8507.36	14631.53	8520.71
	NA2	6368.68	4648.67	6360.95	4655.92	DS3	29265.33	17014.72	29263.06	17041.42
	DW6	3390.38	3390.00	3388.34	3393.52					
	AR4	6295.29	G	6300.85						
	DW7	832.38	832.38	N/A						
	CI1	29265.33	17014.72	29263.06	17041.42	CO1	29083.88	17311.83	17311.83	
	NA3	1097.62	1097.20	1097.24	1097.74	CO2	1254.76	1206.50	1254.76	
	DW8	100.58	100.58	89.36	90.85	CO3	124.89	116.72	181.89	181.69

Of course, the amount and quality of Pu contained in the same spent nuclear fuel will vary according to the difference in burn-up in the nuclear reactor, but the amount of material added or the amount of material produced according to the amount of U and Pu is the ratio according to the chemical reaction formula. The model was built to adjust. Assuming the spent fuel with the composition shown in Table.3, the changes in products and wastes due to facility operation can be estimated as shown in the picture. [5]

Table 3. Input data (Composition of spent nuclear fuel)

Element	Amount(kg)
U	907.1847
Al	29.9824
Si	0.58967
Pu	0.51

If we look specifically at the production of Pu, it shows a production capacity of 0.131924 kg/day when operated without downtime.

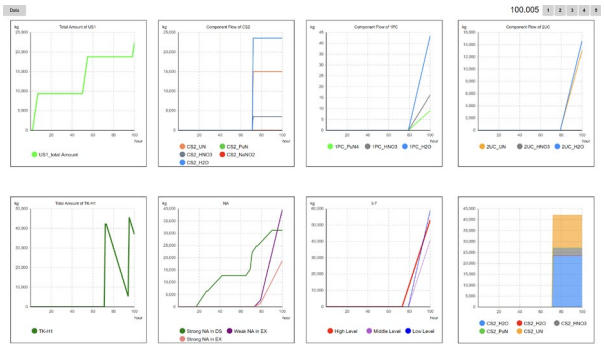

Figure 5. Results of simulation

Acknowledgement

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