

SCRAP RECOVERY PROCESS FOR OXIDE FUEL PLATES

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ABSTRACT

SCRAP RECOVERY PROCESS FOR OXIDE FUEL PLATES. The existing of scrap products in the form oxide fuel plates is very important to be recovered and inputted in the process line. In this paper work, there were 720 scrap of fuel plates (equal to 44,858 grams U) from fabrication process have been dissolved. From this recovery process is obtained 43215.0 grams U in the form dirty oxide fuel powder. Around 27,188.3 grams of U in the form of oxide fuel powder have been dissolved by HNO₃ concentrated. From this dissolution moreless 23,364 grams U in the form of uranyl nitrate have been obtained temporarily. The extraction and stripping process to obtain the purity of uranyl nitrate also has been done. From this process the uranium results in the form uranyl nitrate are 17,563.13 grams. The contents of impurity level of each element also have been compared to the standard limits used in manufacturing the fuel element for research reactors. The total boron equivalent is 3,083 ppm, which is lower than limit of specification.

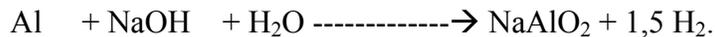
INTRODUCTION.

The plant to produce nuclear fuel element for research reactors has been operated since 1987 to supply fuels for RSG-GAS research reactor. In May 24, 1996, the asset of the plant has been transferred to state owned company (PT Batan Teknologi). During ten years production the plant has shown a good performance to produce fuel element in high quality standard and conforms to the international standard used by other manufacturer in the world. The rejection rate per batch process has been reduced from time to time. There were around 1200 oxide (type of U₃O₈ -Al dispersion) fuel plates rejected after 10 years operation and these fuel plates were an expensive uranium material (LEU less than 20%). To anticipate the uranium to be recovered from fuel plates rejection, the plant has also been completed with the scrap recovery unit. The reasons to use this unit are to consume economically the uranium as feed material, for nuclear fuel accountabilities, and for the safety. The other reason is to recover the uranium in high purity standard so that the uranium can be inputted to the process line. The product of the scraps would be a difference formed and both of them will be treated in step process as follows:

- a. Releasing the low enriched uranium from the other substances such as aluminum, slag CaF₂ to get the U₃O₈ dirty powder.
- b. The dissolution of U₃O₈ dirty powder to get uranyl nitrate dirty solution
- c. Refining the uranyl nitrate dirty solution in order to uranyl nitrate purity solution
- d. Concentration of uranyl nitrate purity solution as feed material in precipitation process (ammonium uranyl carbonate).

The nuclear purity requirements to the product was very important and the use of the aggressive of chemical agent is a must, so that the equipment's to handle this process shall be stainless steel.

A big portion of the uranium scraps from the fabrication process was in the form of fuel plates and fuel cores. The uranium come from this scraps are covered by aluminum. To remove the aluminum from the fuel plates the first step is to use of the Sodium Hydroxide (NaOH) concentrated reagent. By using this method, all the aluminum would be dissolved meanwhile the uranium still in solid suspension and easy to be filtrated. The chemical reaction for the dissolution is:



A high quantity hydrogen will be released from the reaction and rather difficult to be controlled. To avoid any foam built by hydrogen the additional of NaNO₃ will be needed, so that the hydrogen will be oxidized by nitrate to be NH₃.

PROCESS AND EQUIPMENT.

1. Dissolution Process.

By means of burrel pump a number of liters of NaOH 20% are transferred from the supply tank into the dissolver S1CO1. The dissolver S1CO1 is preheated to the operating temperature of T= 110° C . The plate of U₃O₈-Al scraps is dosed by hand. During the dissolution the S1CO1 dissolver is scavenged by supplementary air fed in at the head of the dissolver in order to attain hydrogen concentrations less than 2% by volume. The charging lock for dosing the scrap is blocked when hydrogen concentrations over about 0.5% by volume.

After the dissolution process is finalized, the heating is switch off. When the dissolution product has cooled down to a temperature of T = about 60° C , the contents of the dissolver are drawn off into the vessel S1BO2 by means of vacuum pump S1VO1. After the insoluble uranium product (as U₃O₈ x H₂O) has settled, the sodiumhydroxoaluminate solution over the uranium product is drawn off by means of the vacuum pump VO1, fed through the filter S1FO1 where the solution is filtered. and finally delivered to the vessel BO1. Residual liquid from the removable lower section of the vessel S1BO2 is then drawn off at the bottom of this vessel through the filter emplaced in it by means of the vacuum pump S1VO1, and is likewise fed into the vessel BO1. The filter residue is washed with demineralized water, which is added through the dissolver S1CO1. After analysis to determine the uranium content, the sodiumhydroxoaluminate solution is conveyed by means of compressed air from the vessel S1BO1 into the liquid radioactive waste collection system 5L. After the lower section of the vessel S1BO2 has been detached, the uranium residue is removed and is filled onto dishes. In this dishes, uranium is annealed to uranium oxide in the muffle furnace S1DO1. The uranium results are transported to the sieving box C2AO3, and finally the uranium oxide is filled into the transport and storage containers CBON.

The uranium oxide in the storage containers CBON can be stored or directly transported back to the dissolver S1CO2 for second step dissolution process. To dissolve the U₃O₈ dirty powder a number of liters of HNO₃ 50% are pumped into dissolver S1CO2 by burrel pump

S1PO2. By means of jacket heating the operating temperature is maintained 80° C. The U₃O₈ dirty powder is dosed in from the transport and storage container CBON by the manually operated flap. During the dissolution process, the contents of the dissolver are circulated by compressed air supplied mammoth pump. After cooling to 60° C the contents of the dissolver is drawn off by vacuum pump S1VO1 through the filter S1FO1 and is fed into the vessel S1BO1. After the analysis of the uranium content, the UN solution (filtrate) is transferred to the extraction unit S2 for further process.

2. Extraction Process.

Uranyl nitrate solution from the previous step is dosed into the head of the extraction column S2K01 from the vessel S2B01, or S2B02 by metering pump S2P01. In the countercurrent, TBP (room temperature) is dosed into the bottom of the column by metering pump S2P06 from the vessel S2B03 and S2B04. At the head of the extraction column S2K01, TBP loaded with uranium flows by gravity through the filter S2F01, tracers of water carried along with the U-loaded TBP are separated out. Raffinate is draw off at the bottom of the column by metering pump S2P02 and is fed into the vessel S2B08, S2B09 or S2B12. After determination of the U content, the raffinate is either delivered by metering pump S2P05 to the liquid waste collection system 5L.

TBP loaded with Uranium is dosed in at the bottom of the reextraction column S2K02 from the vessel S2B05 by metering pump S2P03. Demineralized water with HNO₃ are delivered by metering pump S2P07 from vessel S2B06 , S2B07 and S2B11 to the preheater S2W02, heated to 60° C, and dosed into the head of reextraction column S2K02. The uranyl nitrate high purity solution is drawn off at the bottom of the re-extraction column S2K02 by metering pump S2P04 and is fed to the evaporation unit 5S3 to get high uranyl nitrate concentration. TBP flows off the head of the column is cooled to the room temperature in the cooler S2W01, and is collected in the vessels S2B03 and S2B04. From this step process the uranyl nitrate high purity and concentrated can be obtained and ready to go to the next step of Ammonium uranyl carbonate precipitation unit.

3. RESULTS AND DISCUSSION.

3.1 The Dissolution Process

3.1.1 Fuel Plate Dissolution.

The results reported here are based on the process done from May 1998 to July 1999. There were 720 scrap oxide fuel plates containing 44,858 grams of LEU less than 20% that have been dissolved in this period. The results can be seen in the Table 1.

From the Table 1, is shown that the process to recover the uranium from scrap oxide fuel plates has spent the time. The problem with the formation of the foam and gelatine during the dissolution is a difficult one to be controlled during filtration. For 720 plates that have been dissolved partially, the uranium that can be recovered in the form of dirty oxide powders are **43,215.0** grams. And uranium in the form of filtrate is **393.074** grams. The rest of uranium around **1,249.926** grams is still in the inventory process.

3.1.2 Dirty Oxide Powder Dissolution.

By using typical equipment, the oxide powders as a results from the fuel plate dissolution will be treated moreover to get the dirty uranyl nitrate . In this report only 27,188.3 kgs of the uranium results from 3.1.1 have been finally processed into uranyl nitrate. From Table 2 is shown that the result of uranyl nitrate is 23,364 grams U and uranyl nitrate, as product inventory is 3,824.3 grams.

3.1.3 Extraction of Uranyl Nitrate

The next step to process the results of dirty uranyl nitrate is the extraction. The uranyl nitrate results from 3.1.2 are sent to the extraction unit where the uranium will be contacted with Tri buthyl phosphate (TBP) as organic phase. In this process the 30% of TBP is used to purify the uranyl nitrate. More over the results of uranium in the organic phase will be stripped (in the reextraction unit) to get the uranyl nitrate pure relatively. The results of the UN stripping are shown in the Table 3. The purity of the uranyl nitrate from this unit should be tested to determine whether those uranyl nitrate are acceptable for the next step process. The purity of the uranyl nitrate is shown in the Table 4.

CONCLUSION

From the step process to dissolve the scrap oxide fuel plates we could conclude the following:

1. There were 720 scrap oxide fuel plates containing around 44,858 grams U of LEU less than 20% have been dissolved. From 720 plates that have been dissolved partially, the oxide fuel powder around 43,215.0 grams of U can be recovered.
2. The dissolution process to get the uranyl nitrate from oxide fuel powder also has been done. From 27,188.3 grams of U in the form of oxide fuel powder that have been dissolved around 23,364 grams of U in the form of uranyl nitrate are obtained.
3. The extraction and stripping process to obtain the purity of uranyl nitrate also has been done. From this process the uranium results in the form uranyl nitrate are 17,563.13 grams. The contents of impurity level of each element also have been compared to the standard limits used in manufacturing the fuel element for research reactors. The total boron equivalent is 3,083 ppm, which is lower than limit of specification.

REFERENCES

1. Susanto BG, Suropto A, “ Produksi Elemen bakar Reaktor Riset”, Pusat Elemen Bakar Nuklir, 1984.
2. Anonym, “Process Description Dissolution, Filtration, Annealing 5S1”, Revision 1, Volume 4, Contract BATAN – NUKEM, 1983.
3. Anonym, “ Process Description Extraction 5S2”, Revision 1, Volume 4, Contract BATAN –NUKEM, 1983.
4. Laucht J, Mueller H., “ A New Standard For Uranium Metal Intended For Research Reactor Fuel Fabrication”, 21st RERTR Meeting, 1998.

TABLE 1
DISSOLUTION PROCESS FOR OXIDE FUEL PLATES

NO.	Date	NaOH (KG)	DW (ltr)	NANO3 (KG)	T (°C)	Time (t), hour	Number of Plates	Total U (gram)	U in Dirty U ₃ O ₈ Powder (gr)	U in Process Inventory (gr)	U in Liquid Waste (gr)
1	May 18,98	16	85	0	100	8	9	546.93			
2	June 15,98	0	85	0.5	100	10.5	7	425.25			
3	June 29,98	16	85	0.5	100	14.5	14	849.74			
4	July 1, 98	16	85	0	100	14	10	605.62			
5	July 8, 98	16	85	0.5	80	12.5	25	1519.30			
6	July 14, 98	16	90	0.5	80	12	40	2469.98			
7	July 20, 98	16	90	0.5	80	13.5	50	3019.96			
8	July 22, 98	16	90	0.5	80	13	50	3016.25			
9	August 3, 98	16	90	0.5	80	11.5	50	3034.95			
10	August 20, 98	16	90	0.5	80	6.5	19	1147.57			
11	August 27, 98	0	90	0.5	80	7.0	20	1214.12			
12	Sept 7, 98	16	90	0.5	80	8.0	30	1815.39			
13	Sept. 10, 98	0	90	0.5	80	7.0	21	1271.32			
14	Sept. 14, 98	16	90	0.5	80	8.0	30	1809.81			
15	Sept.21, 98	16	90	0.5	80	8.0	30	1798.30	43215,0	1249.926	393.074
16	Sept.23, 98	16	90	0.5	80	8.0	27	1631.79			
17	Sept. 28, 98	18	90	0.5	80	8.0	40	2409.90			
18	Sept. 30, 98	7.5	90	0.5	80	8.0	30	1808.62			
19	June 25, 99	16	85	0.5	80	7.0	30	1804.20			
20	June 30,99	16	85	1	85	7.5	35	2107.84			
21	July 7, 99	16	85	1	85	7.5	30	1812.41			
22	July 17, 99	16	85	1	85	7.5	32	1922.06			
23	July 19, 99	16	85	0.5	85	7.5	30	1799.23			
24	July 22, 99	16	85	0.5	85	7.5	31	1862.73			
25	July 29, 99	16	85	0.5	85	7.5	30	1805.36			
							TOTAL U,	44858	43215.0	1249.926	393.074

TABLE 2
DIRTY OXIDE POWDER DISSOLUTION

NO.	DATE	HNO ₃ (ltr)	DW (ltr)	Tank Number	T ⁰ (C)	Time (Hour)	Weight of oxide to be solved (gr)	Volume of UN results (ltr)	Weight of UN Results (gr)	Oxide or UN Products Inventory (gr)
1	March 4, 99	40	40	CO2	85	3	9,213	80	9,144	
2	March 11, 99	48	40	CO2	90	3	5,888.3	79.42	5,633	
3	March 17, 99	40	40	CO1	95	3.5	6,968	75.85	4,693	3,824.3
4	March 24, 99	60	25	CO2	85	4	5,110.9	75.64	3,826.68	
TOTAL		188	145				27,188.3		23,364	3,824.3

TABLE 3
THE RESULTS OF URANYL NITRATE
EXTRACTION

NO.	Date	UN Feed			The UN Stripping Results			Raffinate as Liquid Waste (gr)			UN Product Inventory (gr)
		Vol (Ltr)	Con. of U (gr/l)	Total U (gr)	Vol (Ltr)	Con. of U (gr/l)	Total U (gr)	Vol (Ltr)	Con. of U (gr/l)	Total U (gr)	
1	March 23, 99	80	114,30	9144	195	46.76	9,118.2	75	0.15	11.25	
2	March 25, 99	79.42	70,92	5633	62	51.07	3,166.34	43	0.12	5.16	5,774.41
3	July 16, 99	75.85	61,88	4693	60	47.32	2,839.2	41	0.13	5.33	
4	July 19, 99	75,64	50,58	3826.68	43	56.73	2,439.39	45	0.12	5.40	
				23,364.68			17,563.13			27.14	5,774.41

TABLE 4
 CHEMICAL IMPURITIES LEVEL OF THE URANYL NITRATE PRODUCT
 COMPARED WITH LIMIT OF SPECIFICATION

NO.	Name of Chemical Impurities	Limit of Impurities (ppm)	Average of UN Product Impurities from Extraction (ppm)	Boron Equivalent Factor	Boron Equivalent (ppm)
1	Ni	*)	36.96	1.08E-03	0.039916
2	Cd	0.5	1.75	3.1E-01	0.542500
3	Co	3.0	-	8.95E-03	-
4	Fe	*)	347.16	6.50E-04	0.225654
5	Mn	5.0	6.52	3.45E-03	0.022494
6	Mg	50.0	45.05	3.70E-05	0.001666
7	Cr	*)	35.14	8.49E-04	0.030054
8	Cu	20	1.76	8.50E-04	0.001496
9	Na	50.0	54.37	2.5E-04	0.013592
10	Li	5.0	-	1.45E-01	-
11	Ca	50.0	30.08	1.53E-04	0.004602
12	Al	100	-	1.21E-04	-
13	Ba	10.0	-	1.2E-04	-
14	B	4.0	2.20	1.0E+00	2.200000
15	Si	50	21.67	8.10E-05	0.001755
	Be	0.2	-	1.0E-05	-
	F	20.0	-	1.0E-05	-
	K	20.0	-	7.6E-04	-
	V	2.0	-	14.1E-05	-
	P	100.0	-	8.0E-05	-
		<input checked="" type="checkbox"/> = 1500 ppm <input checked="" type="checkbox"/> Fe+Ni+Cr = 500 ppm <input checked="" type="checkbox"/> B = 10 ppm	<input checked="" type="checkbox"/> = 582.66 <input checked="" type="checkbox"/> Fe+Cr+Ni = 419.26		<input checked="" type="checkbox"/> BE = 3.083729

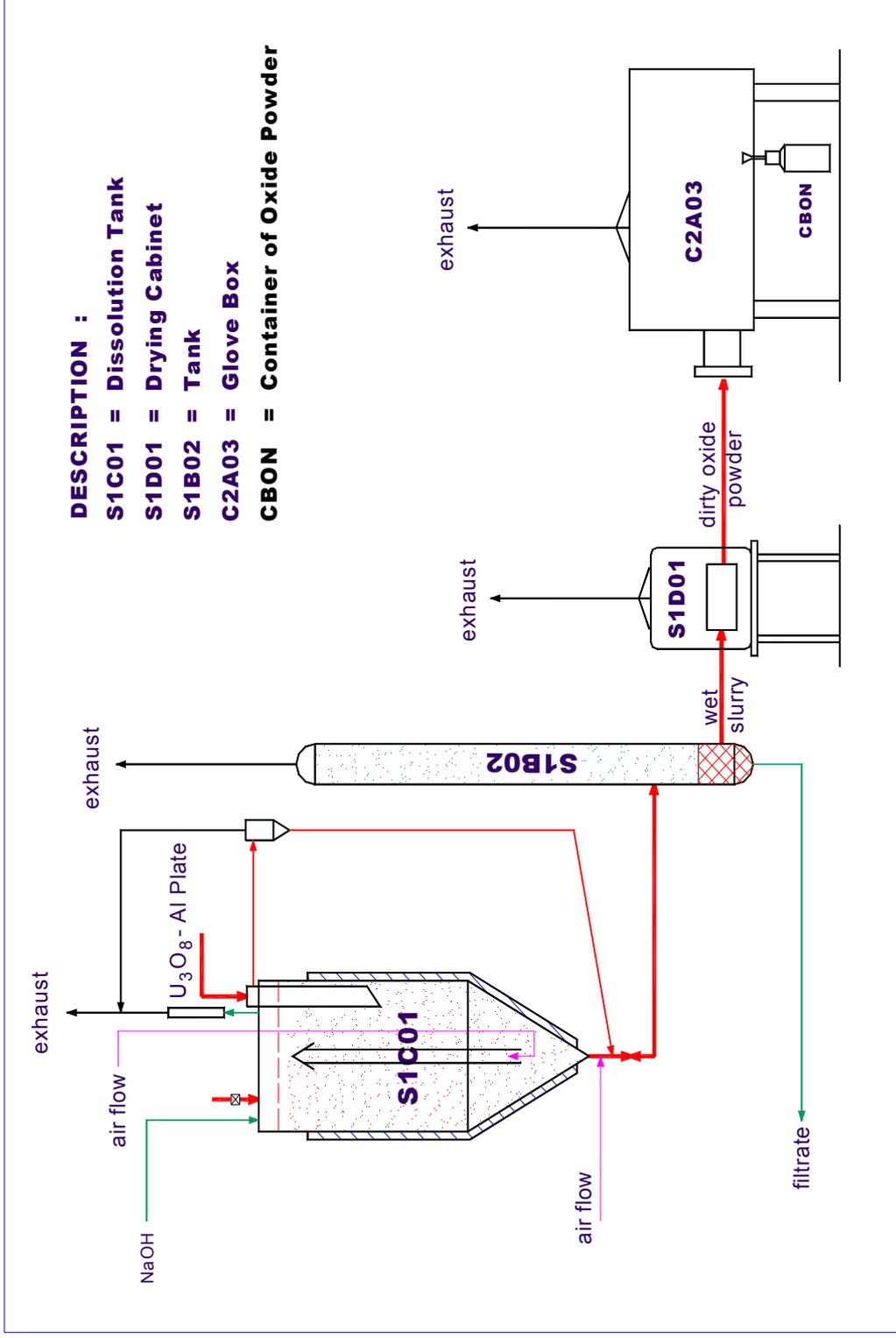


Figure 1: Scrap Recovery Process Flow Diagram for Oxide Fuel Plate

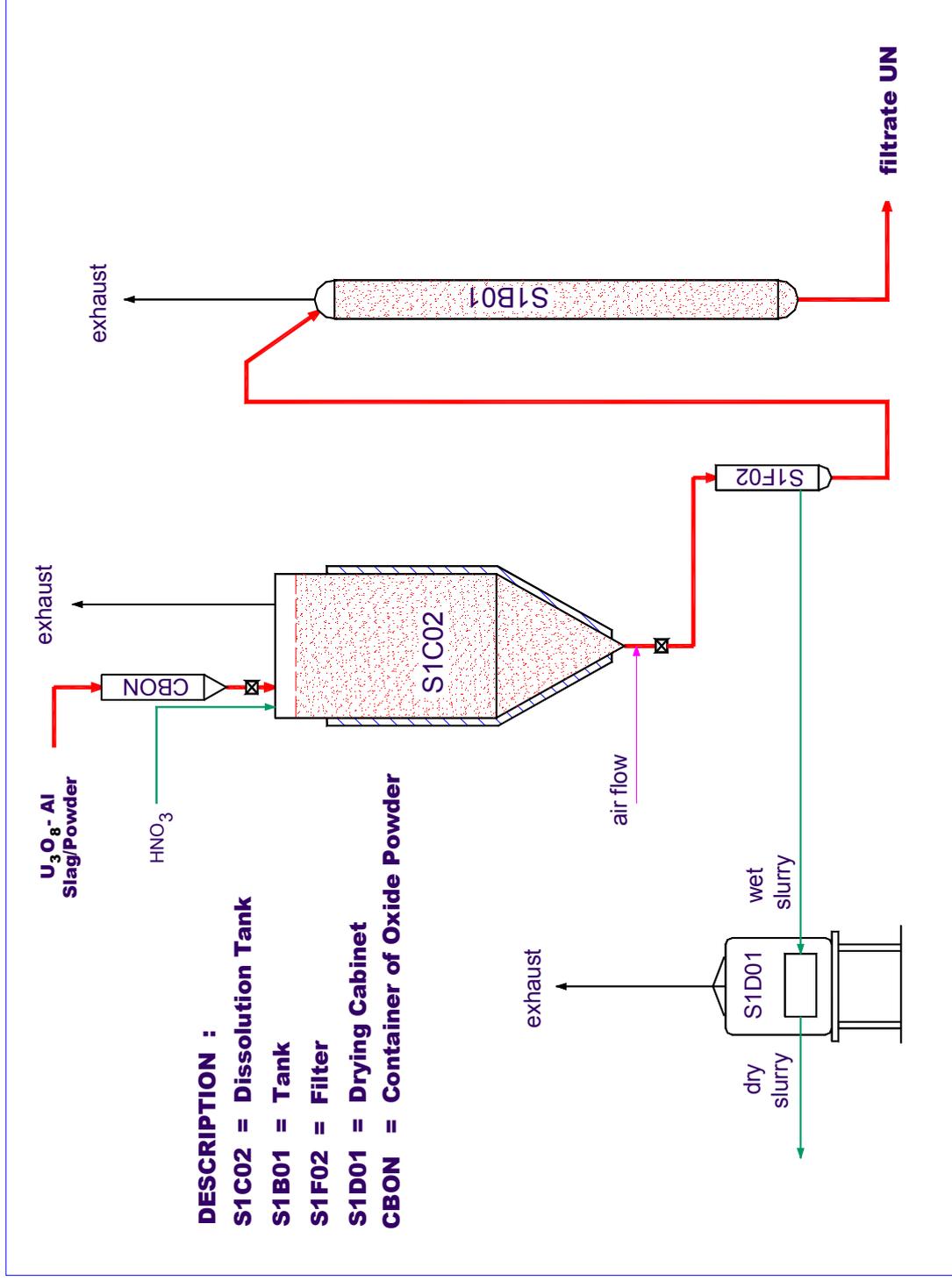


Figure 2: Dissolution Process Flow Diagram for Dirty Uranium Oxide Powder

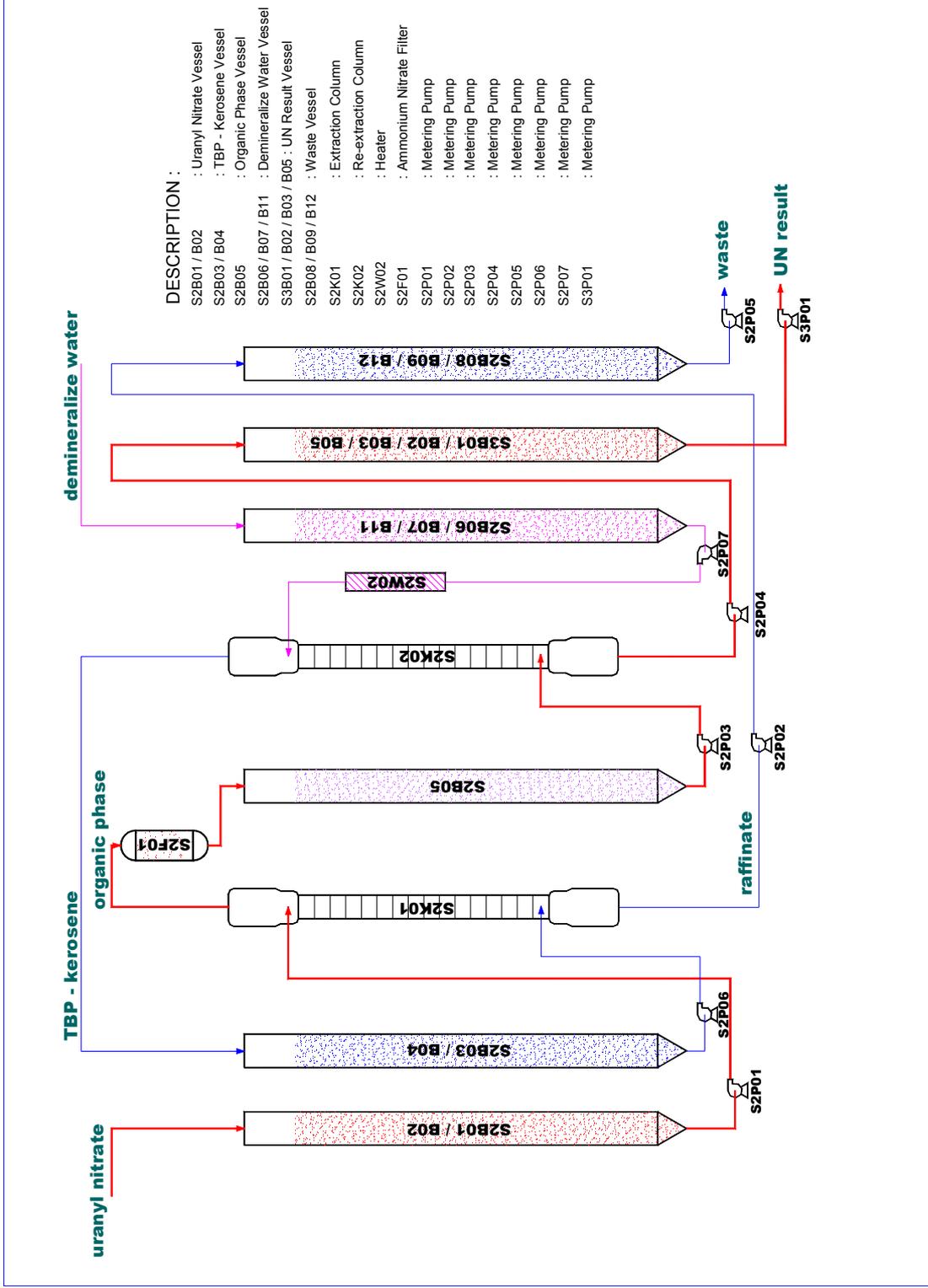


Figure 3: Uranyl Nitrate Extraction Process Flow Diagram