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AEC RESEARCH AND DEVELOPMENT REPORT

PERFORMANCE OF MIXER-SETTLERS IN THE THOREX PROCESS

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Chemical Separations Processes
for Plutonium and Uranium
(TID-4500)

PERFORMANCE OF MIXER-SETTLERS IN THE THOREX PROCESS

by

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Approved by

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February 1966

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ABSTRACT

The capacity of the mixer-settlers in the chemical separations plant normally used for processing enriched uranium is approximately 3000 pounds of thorium per day when using the dilute Thorex process to separate ^{233}U from dissolved thoria and fission products.

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PERFORMANCE OF MIXER-SETTLERS IN THE THOREX PROCESS

INTRODUCTION

A dilute Thorex process has been developed at the Savannah River Laboratory for the recovery of ^{233}U as the oxide and Th as an aqueous solution of $Th(NO_3)_4$ from irradiated ThO_2 or thorium metal. This process is operated in the chemical separations plant normally used for processing enriched uranium. The ^{233}U is separated from the dissolved thoria and fission products by solvent extraction using the seven banks of mixer-settlers presently installed as follows:

First Cycle

1A bank - coextraction of thorium and uranium from fission products with 30% TBP in "Ultrasene"*.

1B bank - back-extraction of thorium with dilute HNO_3.

1C bank - back-extraction of uranium with dilute HNO_3.

Second Uranium Cycle

1D bank - extraction of uranium from residual fission products with 7.5% TBP in "Ultrasene".

1E bank - back-extraction of uranium with dilute HNO_3.

Second Thorium Cycle

2A bank - extraction of thorium from residual fission products with 30% TBP in "Ultrasene".

2B bank - back-extraction of thorium with dilute HNO_3.

The dilute Thorex process was run successfully in small laboratory-scale mixer-settlers at the Savannah River Laboratory.[1] The hydraulic conditions for the 1C, 1D, and 1E banks were used previously in the separations plant equipment, and consequently required no testing. However, the conditions for the 1A, 1B, 2A, and 2B banks differed considerably from those of previous operations. Therefore, tests were conducted in the 14-stage prototype mixer-settlers at the semiworks to determine the hydraulic operability and mass transfer efficiency of the plant equipment in the four new applications. The results of these tests are presented in this report.

*A refined kerosene product of the Atlantic Refining Company.

SUMMARY

Mass transfer tests at the semiworks to determine the capacity of the plant mixer-settlers under dilute Thorex conditions showed that:

1. The solvent extraction capacity of the enriched uranium chemical separations plant operating under dilute Thorex conditions will be about 3000 pounds of Th per day, limited by the 1B thorium stripping bank and the second thorium cycle 2A and 2B banks.

2. The 1B and 2B banks are limited hydraulically to 3000 pounds of Th per day, both by gross entrainment of acid strip solution in the solvent stream, and by pressure drop of solvent flowing through the banks. Both banks will give satisfactory mass transfer at this rate, providing the 1B bank impellers operate at 500 rpm and the 2B bank impellers at 400 to 450 rpm.

3. The 2A bank is limited hydraulically to 3000 pounds of Th per day by the solvent pressure drop across the bank. The mass transfer is satisfactory at this rate, providing the acid concentration in the feed is maintained at about 2.7M HNO_3 and the impellers operate at 400 to 450 rpm.

4. The hydraulic and mass transfer characteristics of the 1A bank are satisfactory at processing rates equivalent to at least 4000 pounds of Th per day, providing the Th concentration in the feed is at the 0.4M maximum value and/or the acid concentration in the 1AS is increased to 2M HNO_3 for increased salting strength. The bank may be limited to a rate slightly less than 3000 pounds of Th per day if the minimum 0.2M Th concentration in the feed and a 1.0M HNO_3 scrub is used. An impeller speed of 450 rpm is necessary.

5. The interface levels in most of the banks are low due to improperly sized orifices on the impeller nozzles when using the enriched uranium mixer-settlers for the dilute Thorex process. However, the back-mixing of solvent entrained with the aqueous phase should not be severe unless the processing rate is decreased below 1500 to 2000 pounds of Th per day.

6. The endstream entrainment is satisfactorily low for all of the banks at processing rates equivalent to 3000 pounds of Th per day.

DISCUSSION

EQUIPMENT

Two sizes of mixer-settler banks are used in the plant: Type 1A,[2] for 1A, 1B, 1D, 2A, and 2B; and Type 1C[3] for 1C, and 1E. Type 1C is larger and is used as an end-fed bank only. The mixer-settler tested at the semiworks was a full-scale prototype of the Type 1A in the plant, except that it contained 14 instead of 16 stages and did not have the remote connections necessary for the plant banks shown in Figure 1.

Figure 2 shows the mixer-settler details and flow pattern through the stages. A stage is divided into three sections: an aqueous inlet section, a mixing section, and a settling section. The aqueous and organic phases flow cocurrently from the mixing section to the settling section within a given stage, but the flow is countercurrent between adjacent stages. The aqueous solution flows into the inlet section and is pumped into the mixing section by an impeller whose suction tube extends into the aqueous layer. The organic phase flows from the settling section of the adjacent stage through a duct into the top of the mixing section. The two phases are mixed, and discharged through louvers into the settling section, where the phases separate. The impellers are driven by variable-speed motors; all impellers in a bank operate at the same speed.

Because the organic phase is not pumped, a hydrostatic head must be developed to force this phase through the bank; this results in a higher solvent level at the organic inlet end of the bank. The solvent will overflow the plant bank at the solvent feed stage if the average pressure drop per stage exceeds 0.4 inch of solvent.

The mixer-settler impellers pump the aqueous phase through the bank. The interface level in the settling sections is dependent on the speed and orifice size of the impellers and on the flow rate of the aqueous phase. Therefore, the orifice sizes in the following table were specified to accommodate the different aqueous flow rates at opposite ends of the center-fed banks of the enriched uranium process, while allowing operation of the impellers at the speeds required for good mixing in each section.

To change orifices in the plant would require the installation of new impellers; therefore, these tests were conducted with the present orifice diameters even though they were not optimum for the dilute Thorex process.

Mixer-Settler	Orifices in 1-1/4-Inch Impeller Nozzles: Stages	Orifice Diameter, inch
1A	1-7	5/8
	8-9	None
	10-16	7/8
1B	1-8	5/8
	9-16	3/8
2A and 2B	All	None

PROCESS DESCRIPTION

The process streams used in the plant are shown in Figures 3 and 4. Clarified solution from the dissolved thoria targets is adjusted to achieve the desired concentrations of aluminum, thorium, and acid. The concentrations must be carefully controlled to prevent formation of a heavy organic third phase and thorium phosphate gels, and to limit the reflux of protactinium in the 1A bank. Thorium and uranium are extracted in the 1B bank with 30% TBP in "Ultrasene". Decontamination from zirconium-niobium and protactinium is improved by adding trisodium phosphate to the 1AS. The thorium and uranium are partitioned in the 1B bank by back-extracting the thorium with dilute acid.

Thorium from the 1B bank is concentrated about two-fold and adjusted with acid to provide feed for the second thorium cycle, where the thorium is further decontaminated using a flowsheet similar to that of the 1A and 1B banks.

The same process streams that are used in the plant were used in the tests of the semiworks mixer-settler, except that no fission products were present and ^{238}U was substituted for ^{233}U. The average run lasted from 5 to 10 hours depending upon the throughput (approximately 17 complete volume changes). The transfer of thorium and uranium to and from the organic phase was used to determine the efficiency under dilute Thorex conditions.

RESULTS

Conditions and results of the solvent extraction tests are summarized in Table I. The principal conclusions from the tests are reported separately for 1A, 1B, 2A, and 2B banks.

TABLE 1

Semiworks Solvent Extraction Tests for the Thorex Process

All tests were made using a 14-stage plant prototype mixer-settler at 35 ±3°C

1A and 2A banks were center-fed at stage 9, 1B and 2B banks were center-fed at stage 8

Bank →	1A			1B			2A		2B		
Impeller speed, rpm →	450	450	450	500	450	450	450	420	450	400	400
Throughput, tons Th/day →	2.0	1.5	1.0	1.5	1.5	1.0	1.5	1.5	1.5	1.5	1.0
Feed											
Aqueous											
H^+, M	1.07	1.00	0.77	0.27	0.26	0.22	1.94	1.90	0.04	0.04	0.04
PO_4^{3-}, M	0.009	0.010	0.008	-	-	-	0.009	0.009	-	-	-
Aqueous[a]											
H^+, M	0.63	0.50	1.10	-	-	-	1.90	2.70	-	-	-
Al^{3+}, M	0.43	0.40	0.55	-	-	-	-	-	-	-	-
Th^{4+}, M	0.20	0.23	0.36	-	-	-	0.39	0.38	-	-	-
U, g/liter	0.41	0.30	0.64	-	-	-	-	-	-	-	-
Organic											
TBP in "Ultrasene", vol %	31.0	30.0	29.5	29.9	30.3	29.5	29.4	29.8	29.4	29.8	29.8
H^+, M	-	-	-	-	-	-	-	-	0.28	0.34	0.34
Th^{4+}, M	-	-	-	-	-	-	-	-	0.09	0.11	0.11
Organic[a]											
H^+, M	-	-	-	0.15	0.16	0.12	-	-	-	-	-
Th^{4+}, M	-	-	-	0.11	0.10	0.12	-	-	-	-	-
U, g/liter	-	-	-	0.17	0.22	0.22	-	-	-	-	-
Endstreams											
Aqueous											
H^+, M	0.38	0.44	0.81	0.50	0.44	0.38	1.22	1.70	0.75	0.75	0.75
Al^{3+}, M	0.45	0.33	0.49	-	-	-	-	-	-	-	-
Th^{4+}, M	0.18	0.20	0.00012	0.18	0.16	0.17	0.060	0.0028	0.21	0.21	0.21
U, g/liter	0.00005	0.0075	0.00026	0.0037	0.0010	0.0023	-	-	-	-	-
Organic											
H^+, M	0.14	0.15	0.13	0.02	0.03	0.03	0.27	0.34	-	-	-
Th^{4+}, M	0.10	0.12	0.13	0.00044	0.0026	0.00006	0.85	0.110	0.002	0.00013	0.00010
U, g/liter	0.22	0.20	0.24	0.15	0.18	0.18	-	-	-	-	-
Losses to Endstreams											
Aqueous											
Th,[b] %	10.0(07)	9.7(0.7)	0.09(<0.03)	-	-	-	22(2)	1.1(0.1)	-	-	-
U, %	0.01	(c)	0.06	1.20(0.5)[d]	0.3(0.07)[d]	0.7(0.2)[d]	-	-	-	-	-
Organic											
Th, %	-	-	-	0.44	3.0	<0.06	-	-	0.2	0.1	0.1
U	-	-	-	-	-	-	-	-	-	-	-
Maximum Endstream Entrainment, vol %											
Aqueous in Organic	1.0	1.0	1.7	3.0	2.5	2.0	1.5	1.5	2.5	2.0	2.0
Organic in Aqueous	0.2	<0.1	<0.1	<0.1	<0.1	0.2	<0.1	<0.1	<0.1	0.2	0.2
Avg Pressure Drop Per Stage,[e] inches of solvent	0.29	0.29	0.13	0.25	0.29	0.07	0.36	0.35	0.36	0.31	0.25
Observed Cocurrent Efficiency, %											
Th	72	80	92	95	81	95	82	78	89	88	95
U	89	92	95	-	-	-	-	-	-	-	-

(a) Center feed stream.
(b) Extrapolated loss for plant mixer-settler which has 3 additional extraction stages (16-stage bank fed at stage 8).
(c) Actual uranium loss is not known because 1AX was contaminated with uranium.
(d) Extrapolated loss for plant mixer-settler which has 2 additional scrub stages (16-stage bank fed at stage 8).
(e) Plant mixer-settler will overflow when the average pressure drop per stage exceeds 0.4 inch of solvent.

1 A Bank

The 1A bank was hydraulically operable at rates equivalent to 4000 pounds of Th per day under the most unfavorable conditions; i.e., with the 1AF thorium concentration at 0.2M, the lowest concentration expected during plant operation, and the acid in the scrub stream at a minimum concentration of 1M HNO_3. Under these conditions the flow of 1AF was increased to maintain the specified concentration of thorium in the 1AP. The thorium loss to the 1AW extrapolated to about 0.7% for the number of extraction stages in the plant bank, at processing rates as low as 3000 pounds of Th per day; this loss was higher than expected. Increasing the speed of the pump-mix impellers from 450 to 500 rpm did not decrease the loss. Apparently, the combination of lower salting strength and increased aqueous flow associated with the low feed concentration produces borderline operation of the 1A bank at 3000 pounds of Th per day. Therefore, the plant bank is probably limited to a rate of slightly less than 3000 pounds of Th per day, unless operated at the 0.4M Th maximum concentration, or unless the acid concentration in the 1AS is increased to 2M HNO_3 for increased salting strength.

The interface levels were low in the scrub end of the 1A bank, as shown in Figure 5, but back-mixing of solvent entrained with the aqueous phase should not be severe unless the processing rate is $<$1500 pounds of Th per day.

1 B Bank

The 1B bank was limited hydraulically to 3000 pounds of Th per day by the rapid increase of gross entrainment of acid strip solution in the uranium product stream and by the pressure drop of solvent flowing through the bank. The mass transfer performance of the 1B bank was satisfactory at this rate with an impeller speed of 500 rpm, but at 450 rpm high thorium losses (3%) were observed in the uranium product stream.

To prevent refluxing of ^{233}U in the 1C bank, the aqueous entrainment in the organic endstream leaving the 1B bank cannot be greater than 3 vol %; this limit was not exceeded at 500 rpm.

The interface levels in the 1B bank will be near the middle of the bank at the anticipated processing rates, as shown in Figure 6.

2 A Bank

The hydraulic capacity of the 2A bank was limited to 3000 pounds of Th per day by the solvent pressure drop across the bank. The mass transfer in the 2A bank was satisfactory at this rate, providing the acid concentration in the feed was increased to 2.7M HNO_3. Operating the bank with the acid concentration in the feed at 1.9M HNO_3, as planned on the original flowsheet, gave a much higher loss (~2%) of thorium than expected due to lack of acid reflux. Higher impeller speeds produced excess solvent pressure drop across the bank.

The 2A bank interface levels were low at an impeller speed of 450 rpm and a processing rate of 3000 pounds of Th per day, particularly in the scrub end of the bank, as shown in Figure 7. Throughput rates lower than about 2000 pounds of Th per day may have serious effects on the mass transfer characteristics of the bank due to excessive back-mixing of solvent entrained with the aqueous phase.

2 B Bank

The 2B bank was limited hydraulically to 3000 pounds of Th per day by gross entrainment of acid strip solution in the solvent exit stream, and by pressure drop of solvent flowing through the bank. The mass transfer performance of the 2B bank was satisfactory at this rate with an impeller speed as low as 400 rpm. An impeller speed of 450 rpm did not significantly improve the mass transfer performance of the bank, probably because of the deleterious effect of an increase in solvent back-mixing with the aqueous phase due to low interface levels, as shown in Figure 8. The amount of interstage solvent entrainment in the aqueous phase increased when the processing rate was lowered to 2000 pounds of Th per day; however, the mass transfer performance was satisfactory at this rate with an impeller speed of 400 rpm.

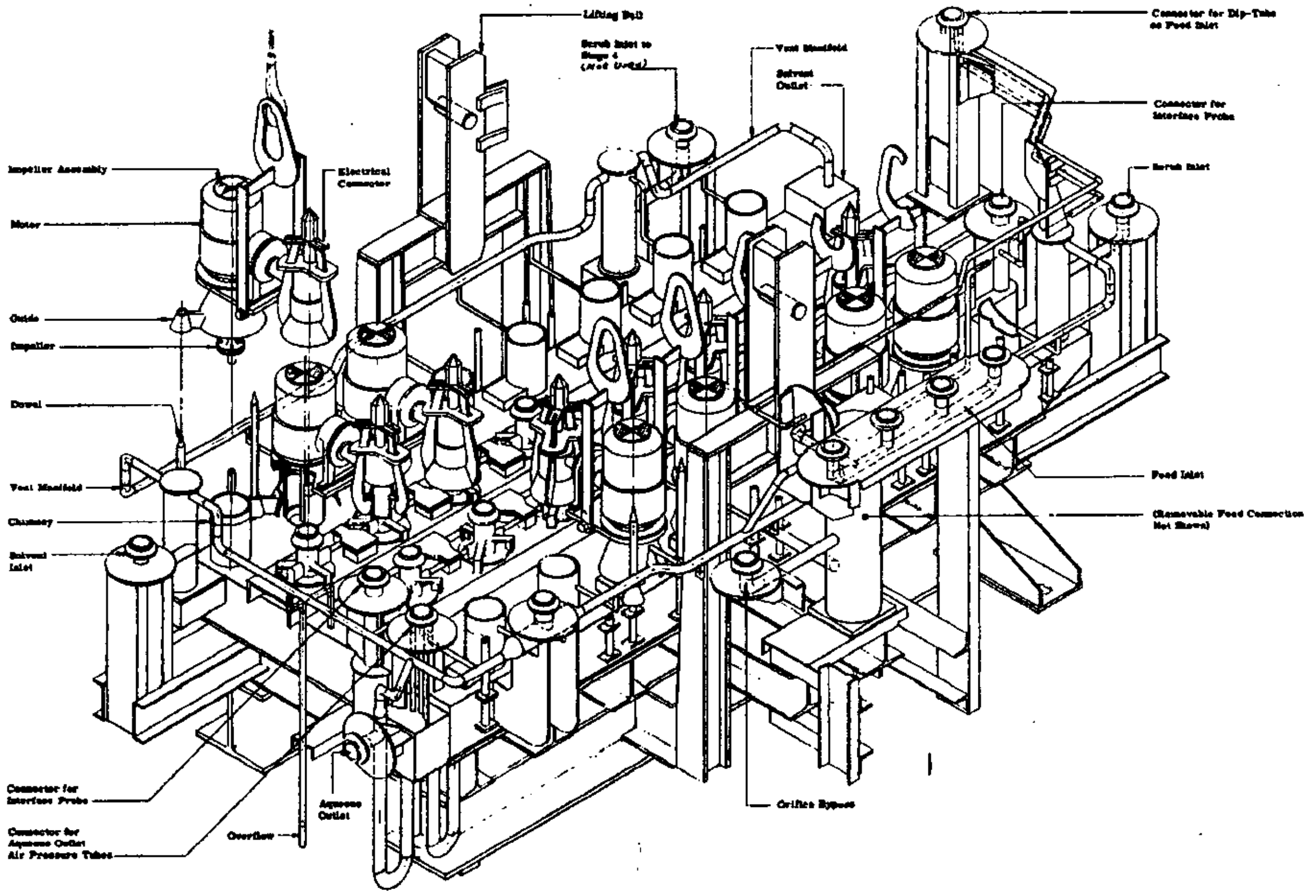


FIG. 1 TYPE 1A MIXER-SETTLER

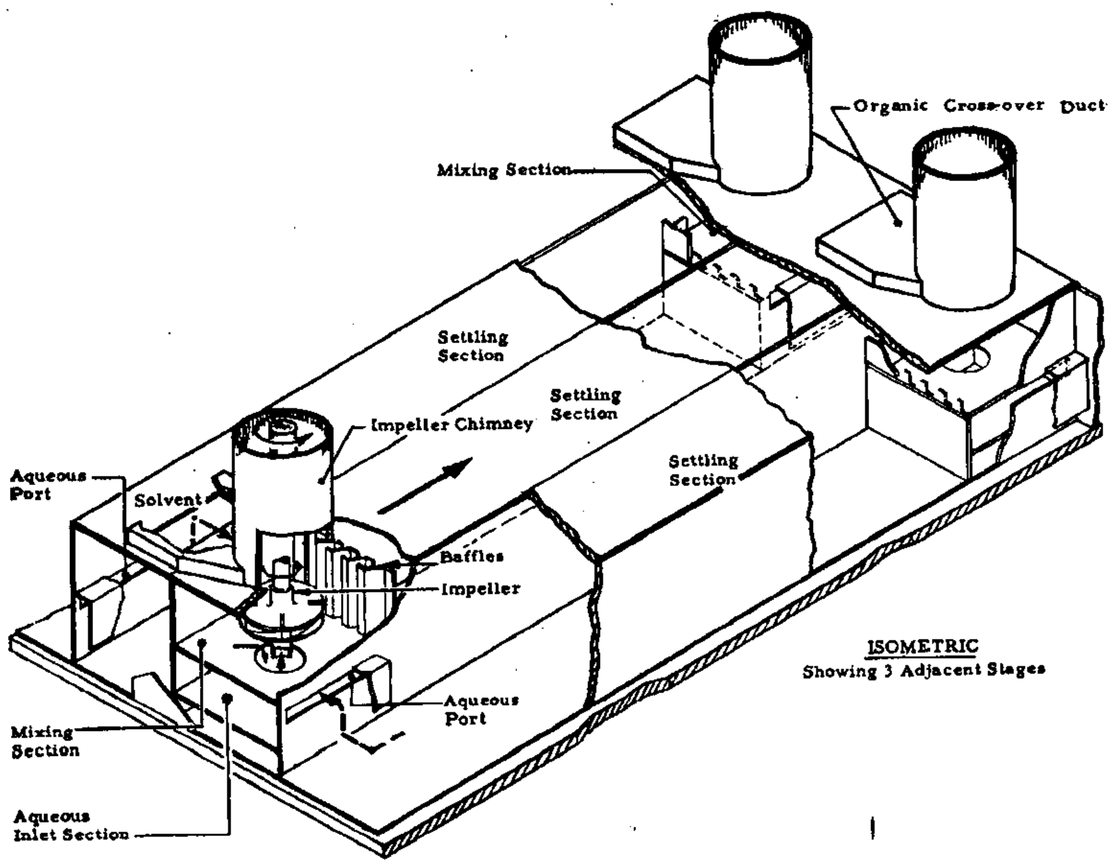
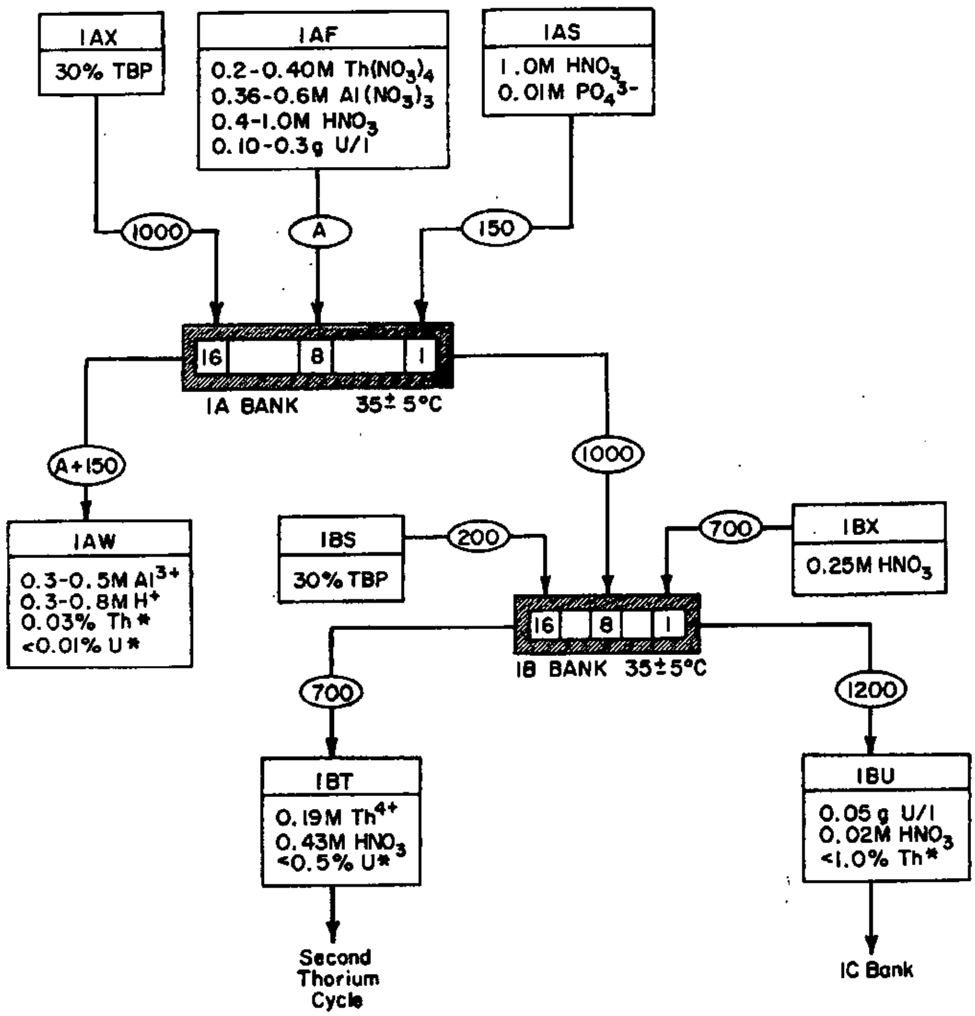


FIG. 2 ISOMETRIC VIEW OF MIXER-SETTLER STAGES – TYPE 1A



(oval) = Relative Flow

*Referred to IAF

A = Relative Flow = 135/ M Th in IAF

FIG. 3 FIRST CYCLE SOLVENT EXTRACTION

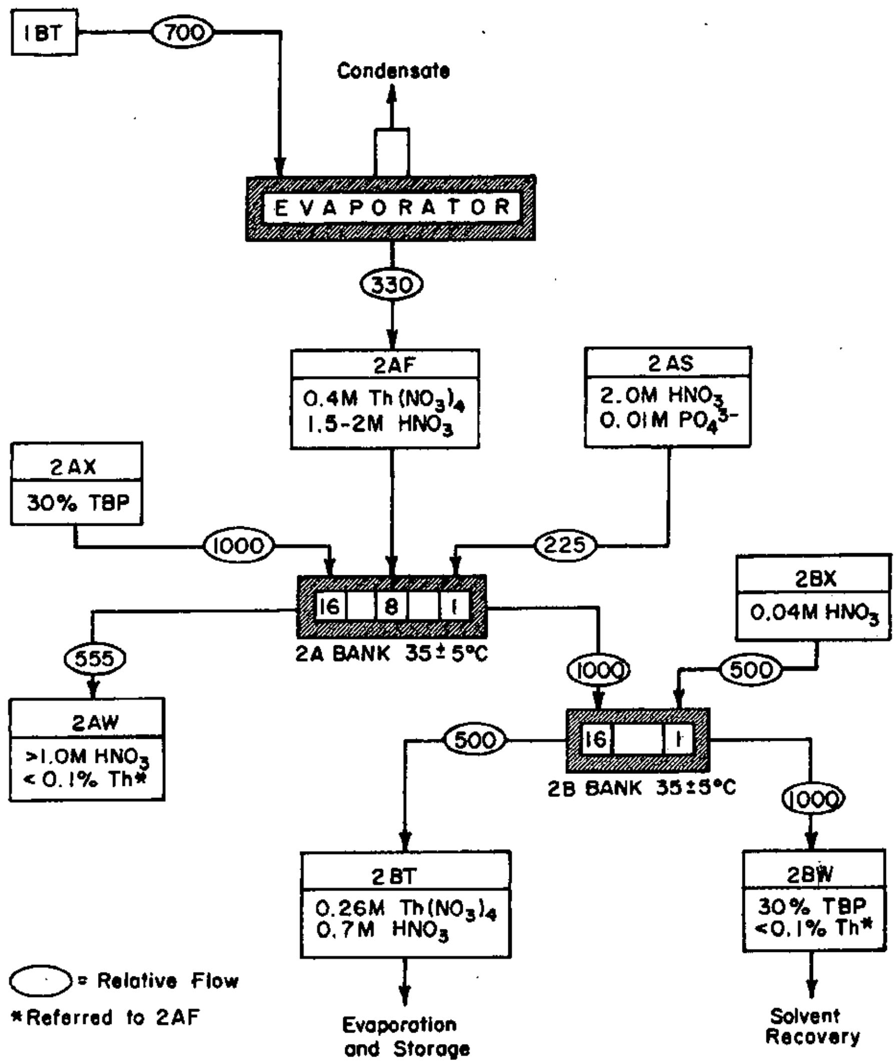


FIG. 4 SECOND THORIUM CYCLE

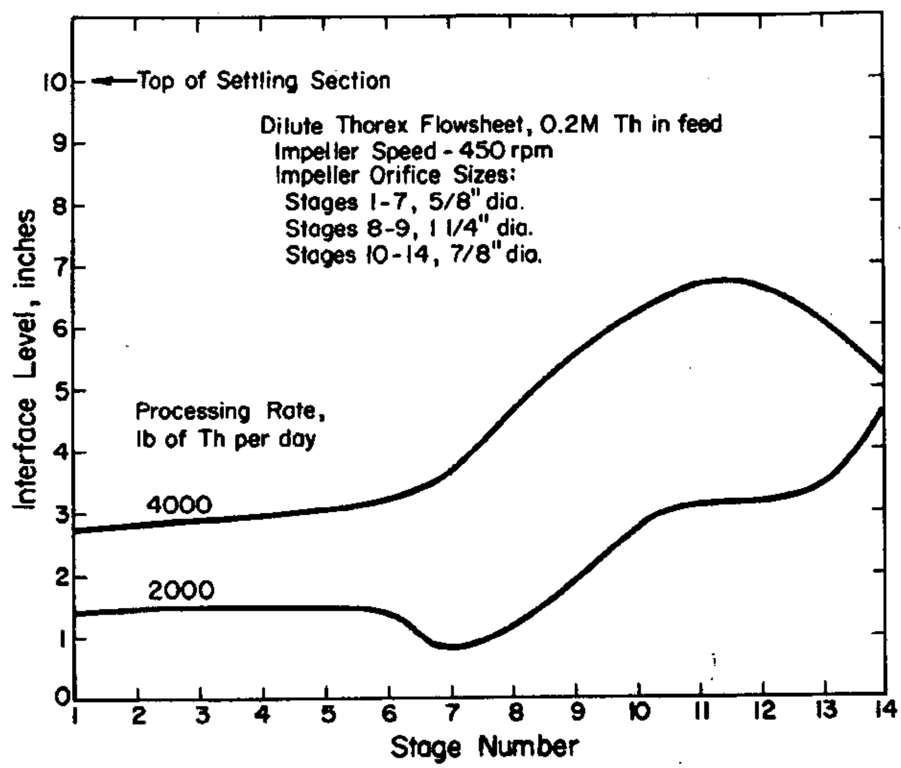


FIG. 5 INTERFACE LEVEL, 1A MIXER-SETTLER

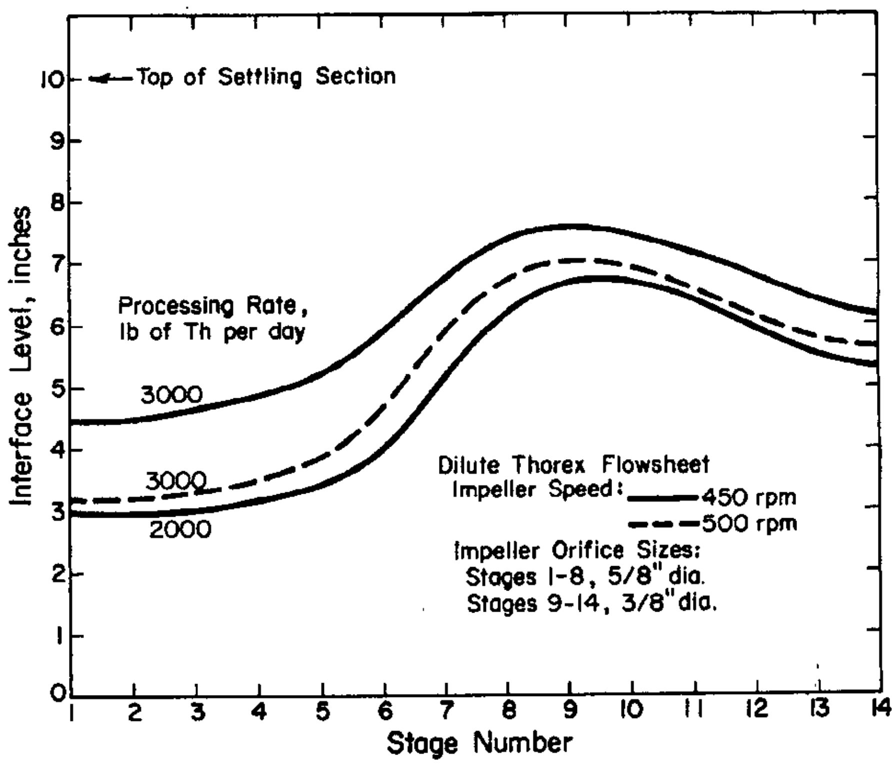


FIG. 6 INTERFACE LEVEL, 1B MIXER-SETTLER

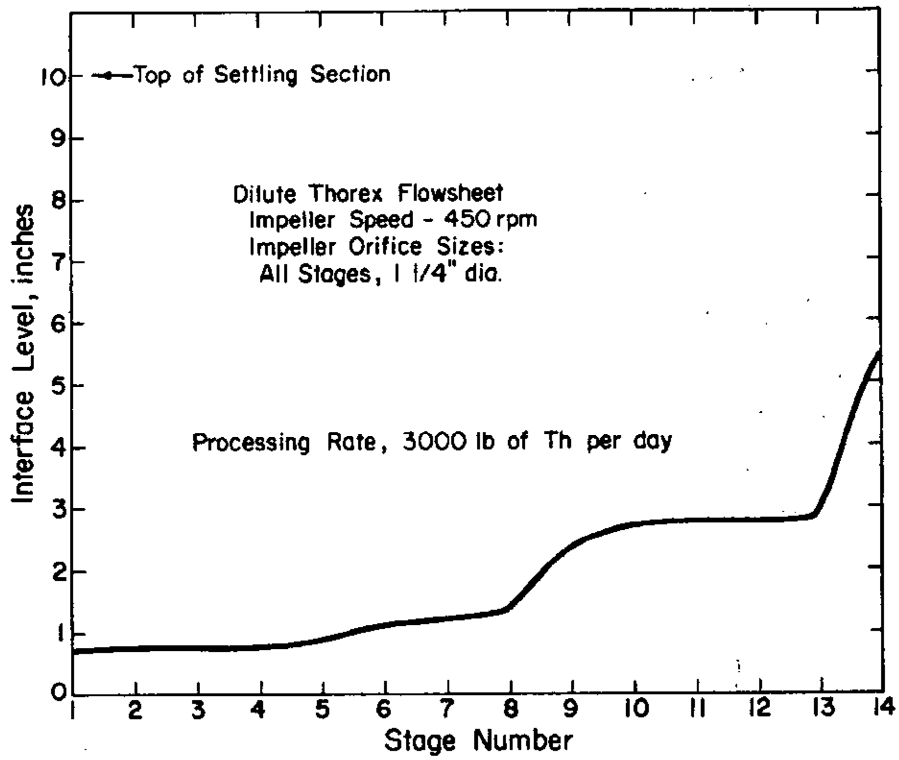


FIG. 7 INTERFACE LEVEL, 2A MIXER-SETTLER

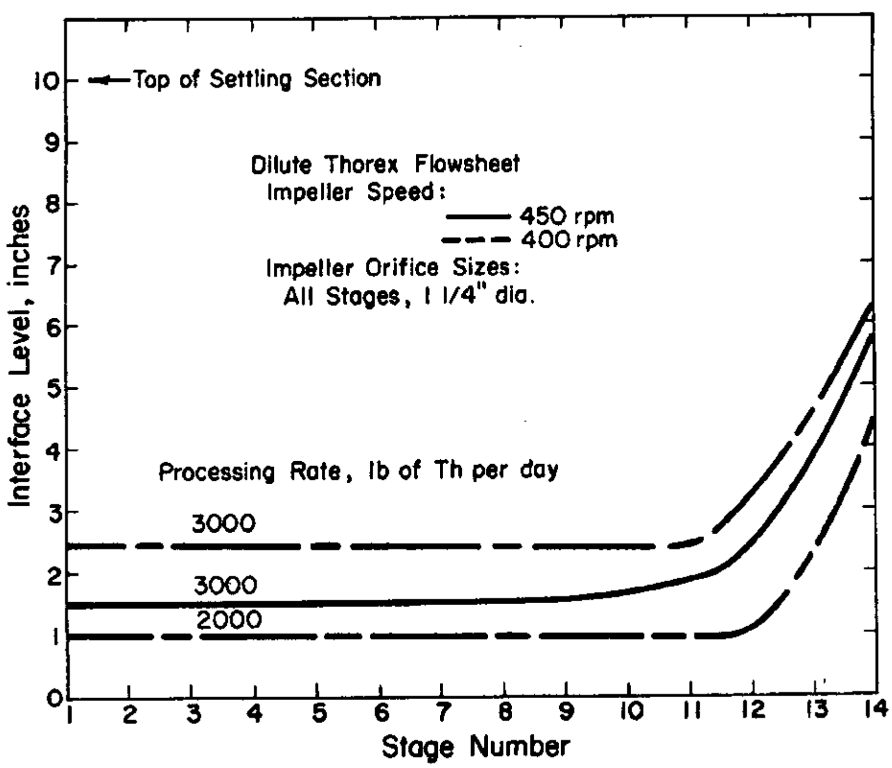


FIG. 8 INTERFACE LEVEL, 2B MIXER-SETTLER

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