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DECONTAMINATION OF THE ORNL  
THOREX PILOT PLANT

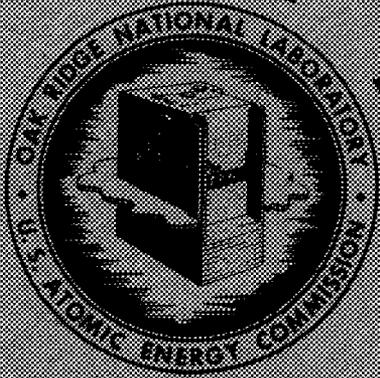
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CHEMICAL TECHNOLOGY DIVISION

PILOT PLANT SECTION

DECONTAMINATION OF THE ORNL THOREX PILOT PLANT

K. H. McCorkle  
W. R. Winsbro

DATE ISSUED

JULY 12 1958

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## 0.0 ABSTRACT

Decontamination of the Thorex Pilot Plant is described. Twenty-four major pieces of highly contaminated stainless-steel processing equipment were decontaminated in 19 days to safe working levels by alternate treatments with 20% sodium hydroxide--2% sodium tartrate and 20% nitric acid. Oxalic acid or citric acid was substituted for tartaric acid in about half the treatments. The average cost per treatment was \$222; the total cost was \$36,000.

## 1.0 SUMMARY

After irradiated thorium metal had been processed in the Thorex Pilot Plant, the plant was shut down and completely decontaminated. In 19 days, the radiation levels from the cell processing equipment, which is housed in stainless-steel-lined concrete- or lead-walled cubicles in three cells, were reduced to safe working levels for maintenance personnel to make equipment changes. After decontamination, the radiation level at the entrance doors to the process cells was 5-10 mr/hr.

After thorium and  $U^{233}$  had been removed from the process equipment, the equipment was decontaminated by alternate treatments with 20% sodium hydroxide-2% sodium tartrate solution and 30% nitric acid. Because of an inadequate supply of tartaric acid, 1% sodium oxalate or 2% sodium citrate was substituted for sodium tartrate in about half the treatments.

The dissolver was decontaminated in 20 treatments. After 10 treatments with caustic-oxalate solution and nitric acid, two heel dissolvings were made with Thorex dissolvent solution (13 M nitric acid), containing excess catalyst, followed by two treatments with caustic-tartrate solution and nitric acid and finally by two treatments with 3% hydrofluoric acid--20% nitric acid alternated with caustic-tartrate. The final radiation level in the cubicle was 300 mr/hr,

most of which was due to localized contamination on the exterior of a tank nozzle through which corrosion specimens were suspended.

The feed preparation, acid recovery, and feed metering equipment was decontaminated to a final radiation level of 150 mr/hr at contact and 75 mr/hr background (about 2 ft away from any equipment in the cubicles) by five caustic treatments alternated with five acid treatments. The extraction column and rework system were also decontaminated to 150 mr/hr at contact and 75 mr/hr background. Four caustic and four acid treatments were required for this equipment. The partitioning column, the strip column, thorium concentrating and storage equipment, uranium product-handling equipment, and the solvent recovery system required four treatments (two caustic and two acid) each to reduce the initial activity level of 2-10 r/hr to 50 mr/hr at contact and 10 mr/hr background.

Analysis of samples showed that protactinium and niobium constituted approximately 80% of the gamma emitters in the effluent decontamination solution. Zirconium, ruthenium, and rare earths comprised, in order of decreasing importance, the remaining 20%. Analysis of samples taken at the beginning and end of decontamination indicated that decontamination of the feed preparation and extraction equipment is limited by protactinium removal; decontamination of the partitioning and rework systems is limited by both protactinium and niobium; and niobium limits decontamination of the stripping column and solvent recovery equipment.

The decontamination equipment, which consists of a solution makeup tank, a decontamination panel (containing a number of quick-disconnect fittings through which the decontamination solutions can be routed to the cell vessels or cubicle sprays), recirculating jets inside the cell tanks, and cubicle sprays, proved to be a worthwhile investment, making possible lower chemical costs, faster decontamination, and lower waste storage costs than is possible with conventional techniques.

Two operators per shift carried out the decontamination program. A total

of 160 treatments (3 hr average time per treatment) was made at a cost of \$222 each. The total program cost was \$36,000. This program was conducted on a development basis. If it were done on a crash basis---3 operators and a shift supervisor on each shift, a minimum of data recording, and only a few samples taken for analysis---the decontamination time and the cost per treatment would be reduced to half these values.

As a part of this program, an attempt was made to study the relative effectiveness of the reagents. The analytical results were too scattered to permit evaluation, but nitric acid and caustic-tartrate are believed to be equally effective, with caustic-oxalate less effective and caustic-citrate poorest.

## 2.0 INTRODUCTION

After the Thorex Pilot Plant at the Oak Ridge National Laboratory had been operated for six weeks, the plant was shut down and decontaminated. The purpose of this was to test the decontamination equipment that had been installed in the plant and to develop and demonstrate decontamination procedures which would result in rapid and significant reductions in radiation levels of cell equipment, such that maintenance personnel would have sufficient working time in the cells to repair equipment failures with a minimum of plant downtime.

Laboratory studies had indicated that 30% nitric acid alternated with 20% sodium hydroxide--2% tartrate should be an effective decontamination procedure. These reagents were selected to be the principal decontaminants for this program, but, because sufficient tartaric acid was not available, oxalate or citrate was substituted in approximately half the caustic treatments. The acid and caustic content of these reagents, when combined on an equal-volume basis, yield a self-neutralized waste, which may be transferred to waste disposal without further pH adjustment.

The entire plant, comprising 24 major pieces of equipment in three cells, was decontaminated. The radiation levels of the equipment before decontamination ranged from greater than 100 r/hr (estimated) to 2 r/hr at contact.

### 3.0 DESCRIPTION OF THE THOREX PILOT PLANT

The Thorex Pilot Plant was constructed in cells 5, 6, and 7 in Bldg. 3019 at ORNL. The plant was designed to be remotely operated and directly maintained. To facilitate direct maintenance, all major equipment pieces were individually shielded (unit shielded) in stainless steel-lined concrete- or lead-walled cubicles easily accessible from a nonradioactive area. All major pieces of equipment and all cubicles housing highly radioactive equipment were designed with built-in decontaminating facilities.

#### 3.1 Building Description

The pilot plant consists of three concrete-shielded cells for housing the radioactive processing equipment, a sample gallery extending the length of the cells on one side, a nonradioactive operating and office area on the other side, an enclosed structure over the cells, and an analytical facility at one end (Fig. 3-1).

Cell 5 is 20 ft square, and cells 6 and 7 combined are 20 ft wide by 40 ft long. Each cell is subdivided into cubicles, each cubicle housing a major piece or pieces of equipment (Fig. 3-2). The cubicles can be entered individually from a nonradioactive (or low-level-activity) area. With this arrangement, only a few pieces of equipment need be decontaminated to be repaired. The floors of all cubicles and the walls of the cubicles in which highly radioactive solutions are handled are lined with stainless steel. All other walls are coated with Amercoat No. 74.

Product  $U^{233}$  is collected in the isolation area. Nonradioactive solutions for the process are prepared in tanks in the makeup area and are pumped from head tanks to the cell equipment. Samples of process solutions are obtained with newly

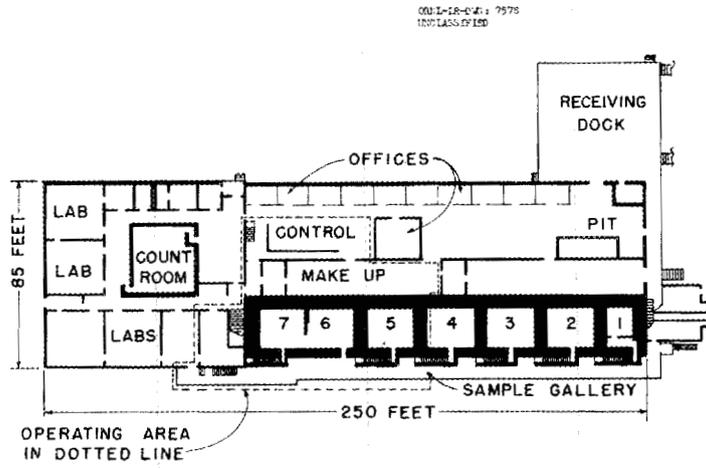


Fig. 3-1  
PLAN OF 3019 BLDG.  
PILOT PLANT

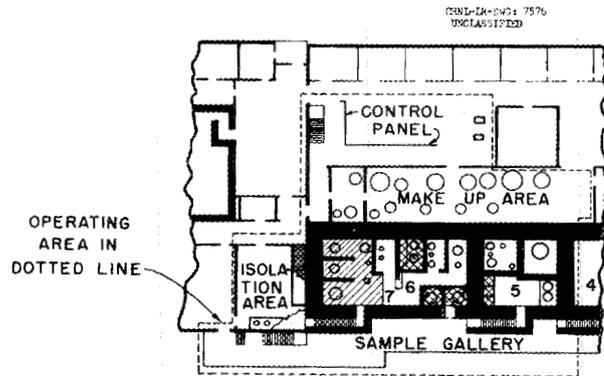


Fig. 3-2  
PLAN OF THOREX OPERATING AREA

developed sampling equipment, which is located in the sampling gallery.

The plant is operated from the master control panel located in the control room. The panel contains indicating, recording, and controlling instruments along with electrical and pneumatic switches for operating the mechanical equipment in the cells, pipe tunnel, and makeup area. The isolation equipment is operated from a panel in that area.

### 3.2 Process Equipment

Over 20 major pieces of processing equipment, such as tanks, columns, evaporators, and a centrifuge, are located in the cells. There are many smaller pieces also, such as phase separators, sampling pots, head pots, jackleg pressure pots, and accumulators. The dissolver and feed adjustment tanks have a capacity of 500 gal each; the remaining tanks range from 150 to 750 gal capacity. The columns are 5 in. dia and 20-40 ft high.

The tanks were constructed with standard flanged dished tops and reverse-dish bottoms. The inlet nozzles (except for an 8-in. flanged inspection nozzle) are of all-welded construction. Each tank has a jacket for steam or water and an internal recirculating jet for decontaminating the interior surfaces.

The process equipment in the cells was constructed from stainless steel, types 309 SNb, 347, or 304L.

### 3.3 Decontamination Equipment

All large tanks are equipped internally with a recirculation jet which picks up decontaminating solution through a suction line from the tank bottom and discharges through two solid-cone spray nozzles against the top of the tank. The dissolver and feed adjustment tank have two jets and four spray nozzles each. The exterior of many tanks and entire cubicles, such as the extraction column cubicle, are decontaminated by spraying solution through solid-cone spray nozzles. The solutions are dropped by gravity from a 290-gal tank in the enclosed area on the roof through a quick-disconnect panel to the desired location.

The jet recirculation spray system (Fig. 3-3) consists of a Schutte-Koerting jet with two solid-cone spray nozzles mounted on the jet discharge. The entire assembly is supported by the steam supply line to the jet, which is welded fast to the flange plate over an 8-in. inspection nozzle on the tank. The unit is compactly assembled so that it can be removed through the 8-in. nozzle. The spray nozzles are located between one-half and three-fourths of a tank diameter down from the top of the vessel. It is not necessary for the conical spray pattern from the spray nozzles to cover the entire top surface of the vessel, since the thrust from the nozzles pushes the reagent out across the top of the tank where it trickles down the walls of the vessel. The jet is located as near the bottom of the tank as possible so that positive jet suction pressures can be maintained with a minimum quantity of reagent. This permits reagent recirculation at solution temperatures up to 85°C.

This system has two advantages over the previous method for effecting internal decontamination of process vessels, namely, that of flooding the vessel with reagent and air sparging for agitation. The efficiency of contact is superior in that complete coverage of the entire upper surface is assured, and agitation at the contact surfaces is much greater because of impingement and trickle effects. The volume of reagent required to effect a single decontamination contact is reduced by a factor of 5-6. This represents a significant reduction in both purchase cost of the fresh reagent and storage cost for the spent reagent.

The 290-gal decontaminating solution makeup vessel is located on top of the cell off-gas duct in the roof area over the process cells. It is equipped with a solution addition funnel, an agitator, a heating coil, and local purge-type liquid-level and specific-gravity instrumentation. (After this program was completed, another tank was installed for nitric acid makeup, permitting uninterrupted solution makeup and drainage to the equipment.) Reagents drain by gravity from the bottom of the tank to the decontamination panel.

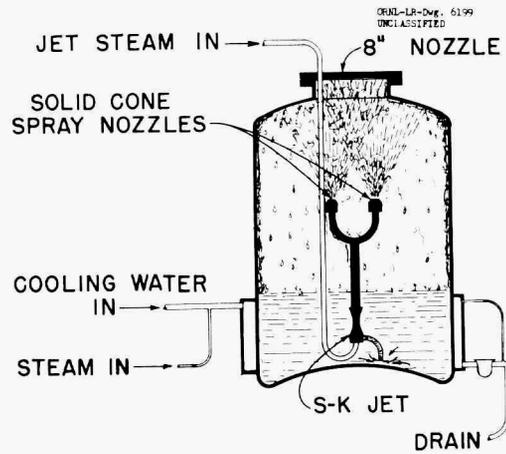


Fig. 3-3  
TYPICAL  
JET RECIRCULATION  
SPRAY SYSTEM

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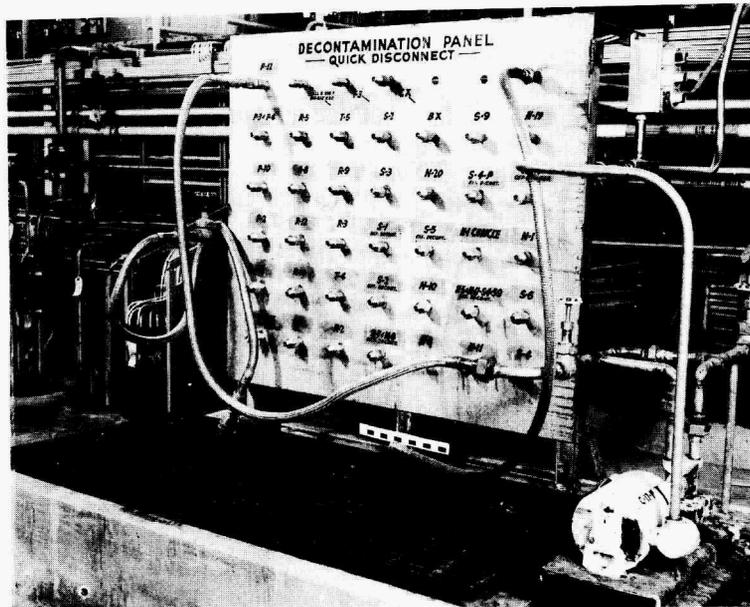


Fig. 3-4  
QUICK-DISCONNECT PANEL

The decontamination panel (Fig. 3-4), located below the decontaminating solution makeup tank, is used for routing decontaminating reagents to various points. Initially during this program, solution was pumped from the makeup tank through stainless steel flexible hoses to the decontamination panel and equipment. Because sufficient head was available for gravity drainage to the cell equipment, the pump was removed and the flexible hoses, which leaked, were replaced with Tygon tubing. The panel now is serviced with caustic and acid solutions from their respective tanks and with steam and water. The Tygon lines are equipped with the socket half of quick-disconnect couplers, while the lines leaving the panel commence with the nozzle half of couplers. The panel couplers are identified according to the destination of the pipes. Reagent routing is effected by mating the socket half of the coupler on the Tygon line with the proper nozzle. Both halves of the coupler contain spring-loaded check valves which seat when the coupler is broken.

#### 4.0 DECONTAMINATION PROCEDURES AND REAGENTS

##### 4.1 Shutdown of the Thorex Pilot Plant

The shutdown of the pilot plant prior to decontamination is an important phase of decontamination, for, if the plant is shut down improperly, significant amounts of valuable materials held up in the system can be lost and downstream equipment can be contaminated excessively. Also, if all thorium is not removed from the system, thorium will be precipitated by the alkaline reagents.

After seven batches of irradiated thorium had been processed in the pilot plant (see Figs. 4-1 and 4-2 for the Thorex feed preparation and solvent extraction flowsheets), the dissolver was rinsed twice with 20 gal of water. The rinses were jettted to the feed adjustment tank, and, after the contents of this tank had been jettted to the feed head tank, the feed adjustment tank was rinsed with two 10-gal quantities of water. These water rinses were combined with the final batch of feed solution prepared from a heel dissolving.

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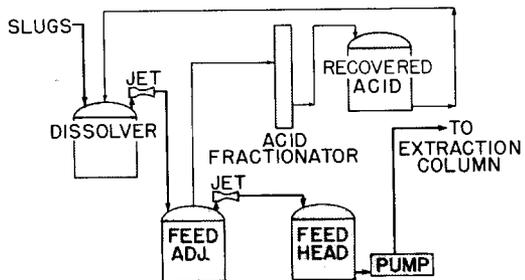


Fig. 4-1  
THOREX  
FEED PREPARATION  
FLOWSHEET

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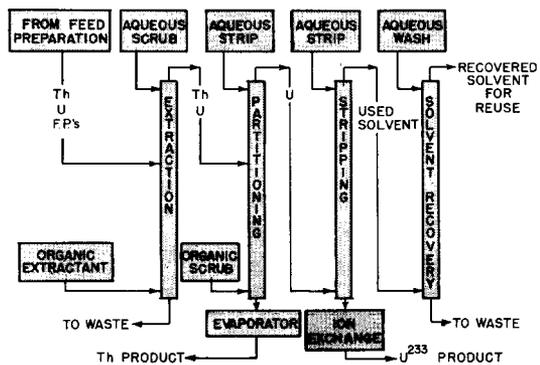


Fig. 4-2  
THOREX  
SOLVENT EXTRACTION  
FLOWSHEET

After all the feed solution had been pumped to the extraction column, two 30-gal quantities of 2 M aluminum nitrate solution were put in the feed head tank and also pumped to the extraction column. The organic stream from the extraction column was sampled every 2 hr and analyzed for gross alpha counts. When the stream contained less than  $10^3$  gross alpha cts/min/ml, the extractant pump was stopped. Aqueous scrub solution was pumped to the column for an additional 4 hr to overflow solvent (including residual  $U^{233}$  and thorium) to the partitioning column.

After the extraction column was shut down, the organic stream from the partitioning column was sampled every 2 hr, and, when the gross alpha count became less than  $10^3$  cts/min/ml, the scrub stream to the partitioning column was stopped. The acid strip solution to the column was diluted in order to decrease the quantity of acid collected in the concentrated thorium catch tanks. Two hours later, the thorium evaporator was shut down and drained. The dilute acid stream from the partitioning column flushed the evaporator for 20 min, after which the column jackleg pressure was increased to stop the aqueous flow to the evaporator. The aqueous stream was allowed to overflow the partitioning column, and when this occurred the aqueous stream pump was stopped. Demineralized water, which had been pumped to the column during this time, was pumped 1 hr longer. The partitioning column was then shut down.

The aqueous stream from the stripping column was sampled and analyzed for gross alpha every 2 hr. When less than  $10^3$  alpha cts/min/ml was contained in the stream, the aqueous flow from the column was stopped and allowed to overflow the top of the column. When aqueous overflow started, the column was shut down.

The solvent recovery column was shut down after the used solvent had been pumped from the solvent storage tank. The carbonate wash stream was allowed to overflow the column momentarily before shutdown.

After this shutdown was completed, all tanks (except those storing product solutions) were drained or jetted. Each vessel having an internal decontamination spray system was steamed for 4 hr.

#### 4.2 Decontaminating Reagents

Decontamination was carried out principally with two reagents, 30% nitric acid and 20% sodium hydroxide--2% sodium tartrate (weight %). These two reagents were selected because: (1) They corrode stainless steel only mildly; (2) they are stable to heating and aging for several days; (3) they are inexpensive compared to most other decontaminating reagents (the cost for 30%  $\text{HNO}_3$  was 5¢ per liter and for caustic-tartrate was 3¢ per liter); and (4) they neutralize one another (equal volumes), allowing convenient waste disposal. In approximately half the caustic treatments, 20% sodium hydroxide--1% sodium oxalate or 20% sodium hydroxide--2% sodium citrate was substituted because sufficient tartaric acid was not available. The concentration of oxalate was decreased to 1% because of the low solubility of sodium oxalate.

Two other reagents were used for decontamination: 10% oxalic acid and 3% hydrofluoric acid--20% nitric acid. Oxalic acid was used in a silica gel column after nitric acid and caustic-tartrate reagents did not reduce the radiation level; 3-20 reagent was used in the dissolver.

#### 4.3 Reagent Contacting Conditions

Alkaline and acid reagents were used alternately. The volume of a batch of reagent was approximately one-third the capacity of the largest vessel in the system being treated. Each batch of reagent was heated to about 60°C and allowed to contact the vessel surfaces for a minimum of 1 hr. In vessels equipped with internal decontamination jets and spray nozzles, the reagent was agitated by the jets. Above 60°C the decontamination jets may fail because of overheating.

Contact times and temperatures could not be controlled in the pulse columns, pumps, and small pots. Several hours was required to fill and empty the concatenated pulse columns, and the large surface-to-volume ratio of the columns and smaller pieces of equipment resulted in cooling of the solutions. Contact time for all vessels except pulse columns began when filling was completed and

ended when emptying was started. Contact time in pulse columns began when filling was completed and ended when the pulser and steam supply to the jacket were shut off.

The acidity or alkalinity of the reagents decreased with increasing contact time and with reuse because of jet dilution and reaction with the tank heel of the previous treatment. Water rinses were not made between treatments.

#### 4.4 Reagent Flow Patterns

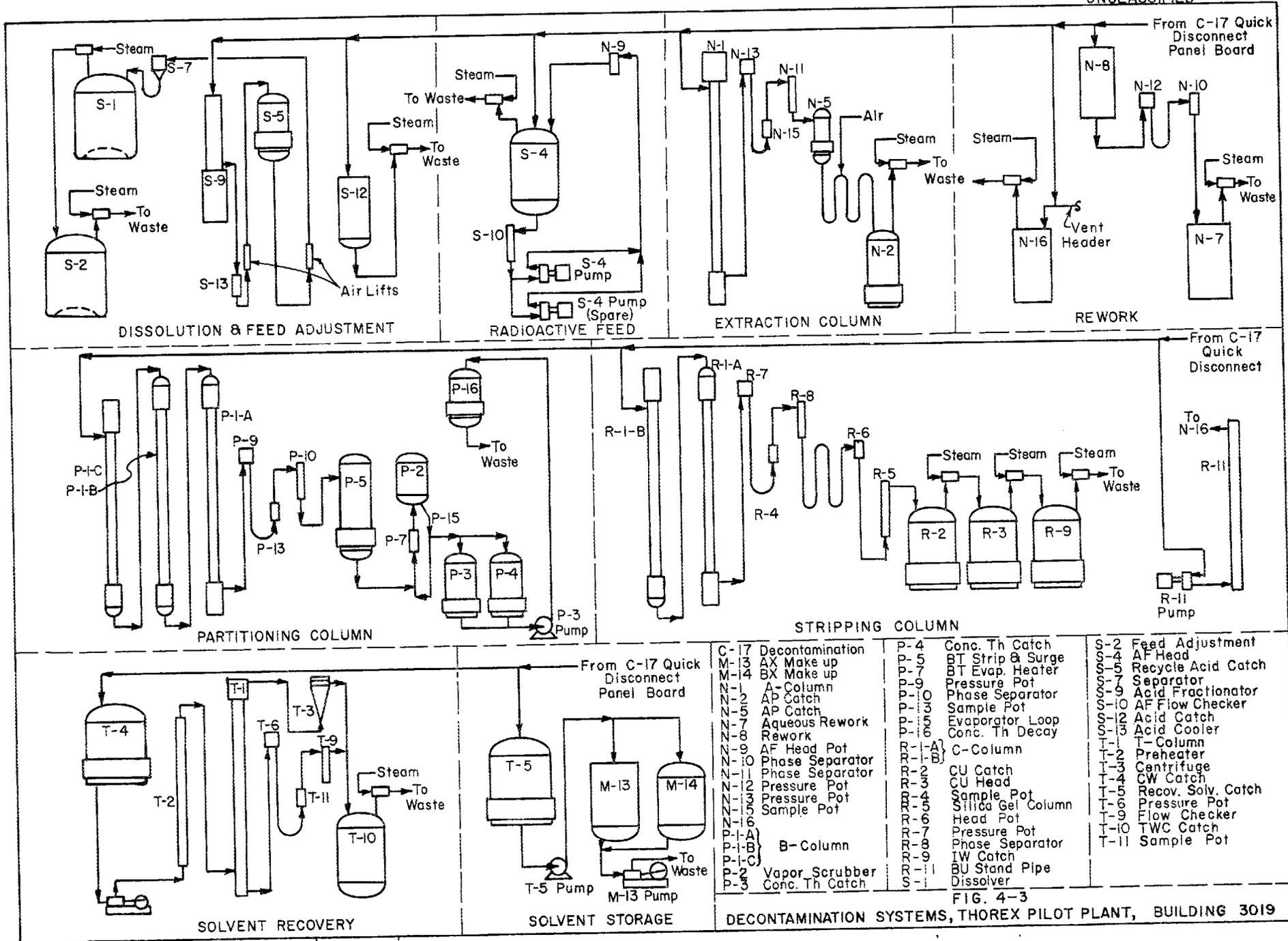
The flow patterns through the equipment were established to obtain the best compromise among the following objectives:

- a. Maximum reuse of reagent
- b. Progression of reagent from vessels of a certain level of contamination to vessels with the same or a higher level of contamination
- c. Simplicity of solution transfer procedures

The equipment was divided into eight groups:

1. Dissolution and feed adjustment
2. Radioactive feed input
3. Extraction
4. Rework
5. Partitioning
6. Stripping
7. Solvent recovery
8. Recovered solvent storage

The pieces of equipment included in each flow pattern and the direction of reagent transfer are shown in Fig. 4-3. Solutions were made up in a decontaminating solution makeup tank, C-17, and were distributed to the various groups via the quick-disconnect decontamination panel. Although generally



C-17	Decontamination	P-4	Conc. Th Catch	S-2	Feed Adjustment
M-13	AX Make up	P-5	BT Strip & Surge	S-3	AF Head
M-14	BX Make up	P-7	BT Evap. Heater	S-4	Recycle Acid Catch
N-1	A-Column	P-9	Phase Separator	S-5	Separator
N-2	AP Catch	P-10	Phase Separator	S-6	Acid Fractionator
N-3	AP Catch	P-13	Sample Pot	S-7	AF Flow Checker
N-4	Aqueous Rework	P-15	Evaporator Loop	S-8	Acid Catch
N-5	Rework	P-16	Conc. Th Decay	S-9	Acid Cooler
N-6	AF Head Pot	R-1-A	C-Column	T-1	T-Column
N-7	Phase Separator	R-1-B		R-1-A	Preheater
N-8	Phase Separator	R-2	CU Catch	T-2	Centrifuge
N-9	Pressure Pot	R-3	CU Head	T-3	CW Catch
N-10	Pressure Pot	R-4	Sample Pot	T-4	Recov. Solv. Catch
N-11	Pressure Pot	R-5	Silica Gel Column	T-5	Pressure Pot
N-12	Pressure Pot	R-6	Head Pot	T-6	Flow Checker
N-13	Sample Pot	R-7	Pressure Pot	T-7	TWC Catch
N-14	Sample Pot	R-8	Phase Separator	T-8	Sample Pot
N-15	Sample Pot	R-9	IW Catch		
P-1-A)	B-Column	R-11	BU Stand Pipe		
P-1-B)					
P-1-C)					
P-2	Vapor Scrubber	S-1	Dissolver		
P-3	Conc. Th Catch				

FIG. 4-3

DECONTAMINATION SYSTEMS, THOREX PILOT PLANT, BUILDING 3019

followed, these flow patterns were not adhered to rigidly. Decontamination also involved side transfers of approximately 20 liters of reagent through each overflow line, the auxiliary jets, and the piping, in addition to the transfers along the principal paths.

External decontamination spray nozzles were used for washing the external surfaces inside the high-radiation-level cubicles.

#### 4.5 Sampling Procedure and Analytical Requests

Samples were taken at the end of all treatments from vessels having samplers. Samples were taken at the beginning of a treatment when it was probable that a significant amount of contamination might be removed in the vessel. Samples were recirculated 0.5 hr during each treatment, and for approximately 10 min at the time of sampling.

Before solutions were transferred to the radioactive chemical waste storage tanks, they were sampled and analyzed for:

1. Thorium
2.  $U^{233}$  alpha or gross alpha (gross alpha was requested if the estimated thorium concentration was less than 10 g/liter)
3. Protactinium gamma
4. Gross beta
5. Gross gamma
6. Ruthenium, zirconium, and niobium gamma and total rare earth beta

Solutions transferred within the system were sampled and analyzed for gross beta, gross gamma, and protactinium gamma.

Requests for hydroxyl ion, hydrogen ion, specific gravity, thorium, and  $U^{233}$  alpha or gross alpha were submitted on a nonroutine basis. Iron was requested only for the metal heel dissolution solutions in the dissolver.

Caustic samples were taken in glass bottles with an interior paraffin coat, and acid samples were taken in glass bottles.

#### 4.6 Radiation Monitoring

Before decontamination was begun, a preliminary survey of the plant radiation levels was made. The radiation from each principal piece of equipment was measured, and the location at which each piece of equipment gave the highest reading was marked. After every two treatments, the radiation levels were measured at the marked locations.

Hard-shell and soft-shell cutie-pies, fish-pole probes, dosimeters, and a specially fabricated small-chambered, long-probe cutie pie were used for monitoring the radiation level of the equipment.

The plant was operated continuously by four rotating shifts, making it necessary for a number of persons to make the radiation surveys.

### 5.0 RESULTS AND DISCUSSION

#### 5.1 Radiation Levels before and after Decontamination

The feed preparation and metering equipment (dissolver, feed adjustment tank, acid recovery tank, and feed tank) was decontaminated from 100 r/hr to 150 mr/hr at contact and 75 mr/hr at background\* (Table 5-1). The intermediate equipment (extraction column, rework system and miscellaneous waste collection tank) was decontaminated from 100 r/hr to 150 mr/hr at contact and 75 mr/hr at background. The radiation levels of the partitioning and stripping columns, the thorium and uranium product handling equipment, and the solvent recovery system, 2-10 r/hr initially, were reduced to 50 mr/hr at contact and 10 mr/hr background (2 r/hr was the radiation level at the base of the partitioning column and 10 r/hr was at the base of the solvent recovery column).

\* Background reading is taken about the middle of a cubicle, 1-3 ft away from equipment.

Table 5-1

Decontamination Treatments and Radiation Levels

Equipment	No. of Treatments	Vol. per Treatment, liters	Radiation Levels		
			Initial, r/hr	Final, mr/hr	
				Contact	Background
Feed preparation and metering	10	900	100	150	75
Intermediate <sup>a</sup>	8	700	100	150	75
Tail end <sup>b</sup>	4	1,800	2-10	50	10

<sup>a</sup>Extraction column, rework system, and miscellaneous-waste collection tank.

<sup>b</sup>Partitioning and stripping columns, thorium and uranium product handling equipment, and solvent recovery system.

The radiation level of three pieces of tail-end equipment decreased rapidly with one acid and one caustic treatment (Table 5-2).

Table 5-2

Monitoring Data for Specific Equipment

No. of Treatments	Radiation Levels, mr/hr		
	Partitioning Column (P-1)	Solvent Recovery Column (T-1)	Used Solvent Storage Tank (T-4)
Before decontamination	2,200	9,000	1,300
After two treatments	100	150	150
After four treatments	65	40	25

The dissolver required treatment with 3% hydrofluoric acid--20% nitric acid followed by 20% caustic--2% sodium tartrate.

### 5.1.1 Dissolution and Feed Preparation Equipment

Except for the dissolver, the feed preparation and acid recovery equipment was decontaminated in 10 treatments, alternating 20% caustic--1% oxalate and 30% nitric acid in the first eight treatments. Caustic-tartrate was used for the ninth treatment (Table 5-3).

The dissolver was decontaminated with the same reagents as the other equipment for 10 treatments, decreasing the radiation level in the cubicle to 4000 mr/hr, after which it was found that undissolved thorium metal was still in the dissolver. Two treatments at boiling with regular Thorex dissolver solution, containing twice the flowsheet catalyst concentration, reduced the radiation level in the cubicle to 400 mr/hr. After two more decontamination cycles (caustic-tartrate followed by nitric acid, repeated twice), and two treatments of 3% hydrofluoric acid--20% nitric acid, which were alternated with two caustic-tartrate treatments, the background in the cubicle was 300 mr/hr. The dissolver was found to have an external area of high radiation near the nozzle in which the corrosion specimens were suspended from the top of the dissolver; this was probably the chief source of background radiation in the cubicle.

### 5.1.2 Feed Metering Equipment

This equipment, feed tank, two Lapp pulsafeeder pumps (one spare), and a pump metering pot, was decontaminated with 10 treatments of 20% caustic--1% oxalate alternated with 30% nitric acid (Table 5-3). The feed tank (500 gal capacity) was decontaminated to a reading of 30 mr/hr.

A Cuno filter in the S-4-P feed pump cubicle and one in the spare pump, S-4-PS, cubicle retained activity and could not be decontaminated sufficiently without disassembly. The S-4-P cubicle background (inside the cubicle) was 3500 mr/hr and the S-4-PS cubicle (pump seldom used) was 200 mr/hr after decontamination. No attempt was made to remove the filters and unnecessarily overexpose personnel. (The filters were removed 7 months later, and, after cleanup, the reading inside the cubicles was 25 mr/hr.)

Radiation Levels and Decontamination Treatments  
of Thorex Pilot Plant Equipment

System	Equip- ment Number	Description	Location	Radiation Level, mr/hr <sup>a</sup>		Reagent Composition <sup>b</sup>											
				Initial	Final	1	2	3	4	5	6	7	8	9	10		
Dissolution and feed adjustment	S-1	Dissolver	S-1 cubicle	100,000	300	0	A	0	A	0	A	0	A	0	A	T	A <sup>c</sup>
	S-2	Feed adjustment tank	S-2 cubicle	100,000	75	0	A	0	A	0	A	0	A	0	A	T	A
	S-5	Recycle acid tank	N-1 cubicle	1,000	80	0	A	0	A	0	A	0	A	0	A	T	A
	S-9	Acid fractionator	N-1 cubicle	-----	60	0	A	0	A	0	A	0	A	0	A	T	A
	S-12	Lean acid tank	Pipe tunnel		90	T	A	T	A	-	-	-	-	-	-	-	-
	S-13	Acid cooler	S-13 cubicle	1,000	30	0	A	0	A	0	A	0	A	0	A	T	A
Radioactive feed input	S-4	Feed tank	N-1 cubicle	100,000	30 <sup>d</sup>	0	A	0	A	0	A	0	A	0	A	0	A
	S-4-F	Feed pump	S-4-P cubicle	11,000	3,500 <sup>e</sup>	0	A	0	A	0	A	0	A	0	A	0	A
	S-4-FS	Spare feed pump	S-4-PS cubicle	2,000	200 <sup>e</sup>	0	A	0	A	0	A	0	A	0	A	0	A
Extraction	N-1	Extraction column	N-1 cubicle	-----	80 <sup>f</sup>	T	A	T	A	T	A	T	A	-	-	-	
	N-2	Waste catch tank	N-2 cubicle	10,000	55	T	A	T	A	T	A	T	A	-	-	-	
	N-5	Waste holdup tank	N-2 cubicle	-----	55	T	A	T	A	T	A	T	A	-	-	-	
Rework	N-7	Rework aqueous tank	Rework cubicle	4,500	60	0	A	0	A	T	A	T	A	-	-	-	
	N-8	Rework tank	Rework cubicle	4,500	200	0	A	0	A	T	A	T	A	-	-	-	
	N-16	Misc. waste tank	N-2 cubicle	-----	55 <sup>g</sup>	-	-	-	-	-	-	-	-	-	-	-	
Partitioning	P-1	Partitioning column	P-1 cubicle	2,200	65	T	A	T	A	-	-	-	-	-	-	-	
	P-3	BT catch tank	P-3 cubicle	1,700	55	T	A	T	A	T	A	T	A	-	-	-	
	P-4	BT catch tank	P-4 cubicle	400	5 <sup>h</sup>	T	A	T	A	T	A	T	A	-	-	-	
	P-15	BT concentrate return	Evap. cubicle	700	10	T	A	T	A	-	-	-	-	-	-	-	
	P-16	BT decay tank	Decay area	900	50	T	A	T	A	T	A	T	A	-	-	-	
Stripping	R-1	Stripping column	R-1 cubicle	700	60	C	A	C	A	-	-	-	-	-	-	-	
	R-2	U product catch tank	Cell 7	500	20	C	A	C	-	-	-	-	-	-	-	-	
	R-3	U product hold tank	Cell 7	800	10	C	A	C	-	-	-	-	-	-	-	-	
	R-5	Silica gel column	R-1 cubicle	3,000 <sup>i</sup>	10	C	A	C	A	Ox	-	-	-	-	-	-	
	R-9	Sorption column waste tank	Cell 7	30	15	A	C	-	-	-	-	-	-	-	-	-	
Solvent recovery	T-1	Solvent recovery column	Cell 7	9,000	40	0	A	0	A	-	-	-	-	-	-	-	
	T-3	Centrifuge	Cell 7	8,200 <sup>j</sup>	8,200 <sup>j</sup>	0	A	0	A	A	Ox	-	-	-	-	-	
	T-4	Waste solvent tank	Cell 7	1,300	25	0	A	0	A	-	-	-	-	-	-	-	
	T-10	Carbonate waste tank	Cell 7	1,000	15	0	A	0	A	-	-	-	-	-	-	-	
Recovered solvent storage	T-5	Recovered solvent catch tank	Cell 7	600	20 <sup>k</sup>	-	-	-	-	-	-	-	-	-	-	-	
	M-13	Extractant head tank	Solvent room	110	10	A	T	-	-	-	-	-	-	-	-	-	
	M-14	P-1 scrub head tank	Solvent room	90	10	A	T	-	-	-	-	-	-	-	-	-	

Footnotes on following page.

<sup>a</sup>The beginning radiation levels for S-1, S-2, and S-4 were estimated. All other beginning levels were measured with a variety of instruments as close as could be gotten to the equipment. Almost all final levels were measured at contact with a perforated paper-chambered cutie pie.

<sup>b</sup>Reagent composition symbols:

A = 30% nitric acid  
C = 20% NaOH---2% sodium citrate  
O = 20% NaOH---1% sodium oxalate  
T = 20% NaOH---2% sodium tartrate  
Ox = 10% oxalic acid

<sup>c</sup>The dissolver was given 10 additional treatments as follows:

11- Thorex dissolver solution with 200% catalyst  
12- Thorex dissolver solution with 200% catalyst  
13- T  
14- A  
15- T  
16- A  
17- 3% hydrofluoric acid---20% nitric acid  
18- T  
19- 3% HF---20% HNO<sub>3</sub>  
20- T

<sup>d</sup>The top of S-4 was surface contaminated to 2000 mr/hr. The background reading near tank S-4 but away from the surface contamination was 30 mr/hr.

<sup>e</sup>Radiation due to a Cuno filter in each cubicle.

<sup>f</sup>Radiation level at the top of N-1 was 80 mr/hr. At the bottom of the column, the level was 250 mr/hr.

<sup>g</sup>Many small transfers from other systems.

<sup>h</sup>Radiation level measured at the top of a barytes block wall which encloses the front of P-4 cubicle.

<sup>i</sup>The silica gel was discarded from the column. Lack of adequate venting caused the decontamination solution to channel in the column.

<sup>j</sup>Shielding reduced the T-3 radiation level to 20 mr/hr.

<sup>k</sup>T-5 was not treated. The apparent decrease in radiation level was due to a reduction in the general background level.

### 5.1.3 Extraction Column Equipment

This equipment was decontaminated with four contacts of caustic-tartrate and four of nitric acid, used alternately. The average radiation level in the N-1 cubicle, which contains the extraction column and associated pressure pot, sample pot, head pot, feed tank, and phase separator, was 75 mr/hr background. A few surface-contaminated spots read 1000 mr/hr. The radiation background in cell 5 doorway was 5 mr/hr.

### 5.1.4 Rework Equipment

Decontamination of the rework system was accomplished by the regular decontamination procedure (eight treatments with caustic-tartrate and nitric acid). After decontamination, the highest activity level found was 600 mr/hr in a localized area near the phase separator.

### 5.1.5 Partitioning Column and Thorium Concentrating Equipment

The partitioning column and thorium evaporator equipment were decontaminated to 40 mr/hr or less with four treatments of caustic-tartrate and nitric acid (two alkaline and two acid). The concentrated thorium catch tanks and decay tank required eight treatments.

Leakage from the seal of a pump (P-3-P, which pumps concentrated thorium solution) caused localized contamination reading 200 mr/hr. Radiation background at cell 6 doorway was 10 mr/hr. The highest general background reading in cell 6 was 40 mr/hr near the sump.

### 5.1.6 Stripping Column and Uranium Product Equipment

Caustic-citrate solution and nitric acid were used in this system. Two to five treatments were required.

The silica gel column remained highly contaminated after five treatments owing to inadequate venting of the column and the use of caustic-citrate solution, which precipitated in the column and caused channeling of the solution through the bed. During a subsequent decontamination after a vent

was installed, one treatment with 30% nitric acid reduced the radiation level of the column from greater than 5000 mr/hr to 110 mr/hr.

The radiation level of R-11-P, which pumps the uranium-bearing organic stream from the partitioning column to the stripping column, remained high at the end of decontamination because of lack of time for a sufficient number of treatments. The pump was shielded with lead brick.

#### 5.1.7 Solvent Recovery System

The solvent recovery system was decontaminated by the general procedure with the exception of the centrifuge, which could not be decontaminated. Most of the radiation from the centrifuge originated in the lower part of the bowl. The radiation level near the bottom of the centrifuge was reduced by wrapping 1 in. of lead around the lower half of the bowl housing and 1/2 in. of lead around the upper half.

#### 5.2 External Decontamination

External decontamination of all exposed equipment was carried out with detergent, water, and sulfamic acid.

The rework system cubicle, N-1 cubicle, S-2 cubicle, and S-1 cubicle were sprayed once with 30% nitric acid and once with water. Analysis of the drainage showed little activity, indicating no need for further decontamination.

A radiation level of approximately 200 mr/hr on the floor under P-4 resulted from draining solutions from P-16 to cell 6 sump. This contamination was removed by two treatments each with 30% nitric acid and 20% caustic-2% tartrate.

#### 5.3 Radiochemical Constituents in the Effluent Activity

The principal radiochemical constituents contributing to the gamma activity were protactinium and niobium, constituting approximately 80% of the total activity. The remaining 20%, in order of decreasing importance, was due to zirconium, ruthenium, and rare earths.

The gross gamma activity in the first decontamination treatments in the feed preparation equipment was about  $10^7$  c/min/ml and in the remaining equipment about  $10^6$  c/min/ml. The gross gamma activity in the final treatments was  $10^5$  c/min/ml in the feed preparation equipment and  $10^4$  c/min/ml in the remaining process equipment. In the first treatment of the feed preparation and extraction equipment, protactinium contributed 10% and niobium 70% of the gross gamma activity. These percentages were reversed in the final treatment; protactinium constituted 70% and niobium 10% of the gross gamma activity. This indicates that decontamination of the Thorex feed preparation and extraction equipment is limited by protactinium removal (Table 5-4). The protactinium and niobium contributions for the partitioning and rework systems (intermediate) and the stripping and solvent recovery (tail end) equipment did not change appreciably from the initial treatment to the final treatment. Both protactinium and niobium appear to limit decontamination of intermediate equipment, while niobium appears to limit tail-end equipment.

Table 5-4

Gamma Activity in the Effluent Reagent

Systems	Gross $\gamma$ , c/min/ml		Percentage of Gross $\gamma$			
			Pa $\gamma$		Nb $\gamma$	
	Initial	Final	Initial	Final	Initial	Final
Feed and extraction	$10^7$	$10^5$	10	70	70	10
Partitioning and rework	$10^6$	$10^4$	30	30	50	50
Stripping and solvent recovery	$10^6$	$10^4$	15	15	65	65

#### 5.4 Decontamination Time and Cost of Decontamination

Twenty-four major pieces of equipment were decontaminated. The total time required for 160 reagent treatments was 480 hr, an average of 3 hr per contact (includes solution addition, heating, cooling, and transfer). This average compares favorably with the actual contact time for all classes of equipment except for concatenated columns, where a contact time of 6 hr is more appropriate. This program was carried out by only half the normal operating force; two operators per shift instead of three operators and a shift supervisor were used each shift. If the normal crew had been used, the total time required for decontamination would have been halved, and the contact time reduced to 1.5 hr.

The total cost of the decontamination program, which lasted 19 days, was \$35,583, as follows:

Labor	\$13,059
Materials	2,117
Analytical services	10,613
Overhead	9,794

The cost per treatment was \$222. The cost for storing 9,000 gal of waste solution is included in overhead.

#### 6.0 CONCLUSIONS

The decontamination procedure recommended is:

1. Dissolve the metal heel in the dissolver with regular Thorex dissolvent containing twice the flowsheet catalyst concentration.
2. Displace the radioactive feed into the extraction column by means of nonradioactive feed until the gamma count (c/min/ml) of the waste stream has decreased by a factor of 1000 and the product stream contains less than  $10^3$  U<sup>233</sup> alpha c/min/ml.

3. After the feed and the extractant streams to the extraction column have been shut off, continue the pulsing and scrubbing until all the organic is pulsed to the top of the column, from which it may be jettted to the rework system for further treatment and storage.
4. After the scrub stream to the extraction column is shut off, allow the partitioning column to operate without feed until the entire input to the BT evaporator is going to the condensate catch tank and the uranium product stream contains less than  $10^3$  U<sup>233</sup> alpha c/min/ml.
5. After the partitioning column has been shut down and the BT evaporator has been drained to the BT catch tank, continue operation of the stripping column until the uranium product stream contains less than  $10^3$  U<sup>233</sup> alpha c/min/ml.
6. Continue operation of the solvent recovery column and the product sorption column until all material in the feed tanks for the columns is exhausted.
7. Shut down the solvent recovery column by setting the jackleg pressure and scrub input rate at maximum until all the solvent in the column has been displaced to the solvent storage system.

The shutdown procedure used prior to this program left thorium and uranium in the dissolver as a result of insufficient cleanout. Activity was carried over from the extraction column to the partitioning column and downstream equipment, and thorium was lost from the partitioning column because caustic solution pumped to the column before all thorium was removed from the column precipitated the thorium. For subsequent decontaminations of the Thorex Pilot Plant, the shutdown procedure should be modified such that activity would be discharged to the waste stream from the extraction column. A modified procedure would also conserve uranium and be less time consuming, but would result in a loss of about 30 kg of thorium which is held up in the extraction column.

Alternate treatments with 20% sodium hydroxide—30% nitric acid provided rapid, economical, and satisfactory decontamination. These two reagents would have sufficed if the dissolver had been thoroughly cleaned of metal heel and if sufficient tartaric acid been available for the program.

The decontamination equipment (internal sprays and jets, external sprays, and disconnect panel) proved to be a worthwhile investment. The required volume of reagent per contact was much less than would have been required without the equipment. The design of the vessels—dished heads and bottoms and all-welded nozzles—also aided in the efficient internal surface decontamination. The decontamination panel with its quick disconnects was handy in adding solution to the cell vessels. If a second decontamination solution makeup vessel had been available, solution makeup scheduling would have been less difficult.

Adequate monitoring equipment is needed for radiation measurements. Because several health physics surveyors monitored the equipment and many different types of instruments were used for measuring the radiation levels, the radiation measurements taken after each treatment of a piece of equipment in many cases could not be correlated directly with other treatments.