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Runs E-3 through E-6
TO: F. L. Culler
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ABSTRACT

Reprocessing of the ARE fuel was resumed after extensive leak testing in the pilot plant. This was considered necessary to assure no recurrence of gaseous UF_6 leaks as experienced in Run E-2. In the four additional runs required to complete the program, about 641 kg of fluoride salt containing 40.64 kg of fully enriched uranium was reprocessed. Recovery as UF_6 product represented 97.97% of the feed, with 0.01% measured losses. An additional 2.14% was reclaimed from NaF beds. The product was of sufficient purity to meet specifications for material designated for reduction to uranium metal. Decontamination from fission products was essentially complete.

Calculations based on the entire ARE program indicated 96.38% product recovery, with 0.06% measured losses. An additional 2.50% was reclaimed from NaF beds and equipment washes.

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1.0 INTRODUCTION

The Fluoride Volatility Process was developed to recover uranium from fused salt reactor fuels. The process consists of volatilization of UF_6 from molten salt by fluorination of UF_4 , and additional decontamination of the UF_6 from volatile or entrained fission product fluorides by sorption and desorption in fixed beds of granular NaF.

A pilot plant scale study was started in December, 1956. Design of the plant was based on a batch process, each batch of salt containing ~ 10 kg of highly enriched uranium. A program of equipment shakedown and process runs with nonradioactive salt has been completed.

To satisfy a production commitment and to obtain experience with tracer level radioactive salt a series of "E" runs was performed in which uranium was recovered from the fluoride salt fuel burned in the ARE. Primary emphasis was placed on uranium recovery, with development data being secondary. After two runs had been completed, processing was interrupted because of a UF_6 leak. The pilot plant was shut down and an extensive leak-test program was initiated. After all faulty connections and seals had been made gas-tight, processing of the ARE fuel was resumed and carried to completion.

This report covers processing of the ARE fuel through the last four runs (Runs E-3, -4, -5, -6) plus a summary of the entire ARE reprocessing program.

2.0 SUMMARY

After extensive leak-testing, the ARE reprocessing program was resumed, and four runs were required to recover the remainder of the salt. Very few operational difficulties were encountered, and consequently the four runs were completed at the rate of two per week (~ 20 kg U/week).

The over-all material balance for the entire ARE reprocessing program indicated a recovery of 59.34 kg uranium (96.38% of feed) as UF_6 in product cylinders. An additional 1.0 kg (1.65%) was trapped on NaF beds, and 0.5 kg (0.85%) was flushed and/or purged from the system; this represented a total recovery of 98.88% of the feed. Measured loss was 34 g of uranium, and 655 g was unaccounted for. The latter item was attributed to the major leak of UF_6 which occurred during Run E-2(1) and an estimated holdup of ~ 50 g in the cold traps.

Because of the aforementioned leak, however, the recovery figures for only the last four runs are more representative of the capabilities of VPP. About 641 kg of salt containing 40.6 kg of fully enriched uranium was reprocessed in Volatility Pilot Plant Runs E-3 through E-6. Of this, 39.8 kg (97.97% of feed) was recovered in product cylinders as UF_6 , 0.5 kg (1.26%) was trapped on NaF beds, and 0.36 kg (0.88%) was purged from the system; this represented a total recovery of 100.12% of the feed. Measured loss was 6 g of uranium (0.01% of feed) in the waste salt. The excess 51 g (0.13%) was within the accuracy of the material balance calculations.

The UF_6 product was of sufficient purity to meet specifications for material designated for reduction to uranium metal. The cation content--notably Cr, Cu, Fe, and Ni--exceeded specifications for product level UF_6 to be returned to a gaseous diffusion cascade.

Decontamination from fission products was essentially complete. Although radiochemical analyses indicated a Te γ concentration of ~ 2 cpm/mg U, the validity of this analysis was considered questionable because of the Gr γ analysis of 8-10 cpm/mg U (U background = 11 cpm/mg U). Gr β activity indicated uranium to be the only β -emitting material present in the product. Thus, gross gamma decontamination factor from fission products of infinity was demonstrated. For comparative purposes a Gr γ activity of 1 cpm/mg U would result in a decontamination factor of 9×10^4 . Apparently, the low activity of the feed salt ($\sim 5 \times 10^6$ Gr γ cpm/g salt) was a limiting factor. About 99.8% of the fission product activity remained in the salt throughout the runs.

From an operational standpoint the final four "E" runs were much improved over previous runs in the pilot plant. The only major difficulty involved waste salt transfer, in which some salt was spilled onto the cell floor. Gross gamma readings of up to 7 r/hr were observed in Cell I-A prior to removal of the spilled salt. Although this operation was never completely adequate, performance was improved in later runs by better heating facilities and operating techniques.

3.0 URANIUM MATERIAL BALANCE

During the last four ARE runs the UF_6 product recovered in cylinders (97.97%), plus holdup (0.88%) and NaF beds (1.26%), represented a total recovery of 100.12% of the total uranium in the feed. Total measured loss was 0.01% of the feed. This excess of 51 g was within the accuracy of the material balance (Table 3.1).

In Table 3.2, the summary material balance for the entire ARE reprocessing program is shown. Total recovery represented 98.88% of the feed, with 96.38% being collected as UF_6 product. Measured loss was 0.06%, and 1.06% (655 g) was unaccounted for. This balance does not include the estimated 768 g of uranium that was lost during the UF_6 leak in Run E-2, ⁽¹⁾ and an estimated holdup of ~ 50 g of uranium in the product transfer system.

Improved operating technique for the CRP trap and longer service life of the absorber beds effectively reduced the percentage of uranium retained on NaF beds from 2.24% in the first two ARE runs to 1.26% in the last four runs. The system holdup listed as 358 g represents the uranium obtained when the system was purged with nitrogen after the last run. Another 50 g to 100 g probably remained in the cold traps, but flushing the system with water was considered undesirable at that stage of the reprocessing program in the pilot plant (Sec. 15.3).

The recovery figures calculated from VPP measurements were checked by Y-12 measurements of the UF_6 and aqueous solutions shipped to that installation. Y-12 accountability measurements were considered the more accurate, and therefore these figures were incorporated in the material balance

Table 3.1 Uranium Material Balance for Runs E-3 through E-6

	Weight of Uranium, g					Total	
	E-3	E-4	E-5	E-6	Post E-6	Wt., g	% of Feed
Feed	10,868	11,294	10,127	8,349		40,638	
Product	10,037	11,183	9,670	8,516	406 ^a	39,812	97.97
Trapped on NaF ^b							
1st Absorber					28		
2nd Absorber					16 ^c		
Complexible Radio-active Products Trap	11	13	194	3			
Main Chemical Trap		14		61			
Vacuum Chemical Trap		147		19			
Heated Duct Chemical Trap					7		
TOTAL						513	1.26
System Holdup ^d							
Purge (FV-124)						358	0.88
Loss, Waste Salt	1	3	1	1		6	0.01
Accounted For						40,689	100.13

a. Y-12 measurements on product received showed an excess of 340 g U. An additional 66 g was washed from the empty product cylinders upon return to VPP.

b. Recovery was made by dissolution of the NaF pellets and subsequent aqueous processing.

c. 23 g U found on top layer of NaF was attributed to "system holdup" because of an accidental flow reversal during purging operations.

d. System was not washed, only purged with nitrogen after Run E-6. An additional 50-100 g U probably was retained in the cold traps.

Table 3.2. Summary Uranium Balance for the Entire ARE Program

	Weight of Uranium, g	% of Feed
Feed	61,629 $\pm$ 770	
Product (UF ₆)	59,397 ^a $\pm$ 80	96.38
Trapped on NaF ^b	1,018 ^c $\pm$ 102	1.65
System Holdup ^d	525 $\pm$ 52	0.85
TOTAL RECOVERED	60,940 $\pm$ 140	98.88
Measured Loss	34	0.06
Accounted For	60,974 $\pm$ 780	
Unaccounted For	655	

- a. Includes 340 g excess at Y-12 plus 105 g washed out of the returned cylinders.
- b. Pertains only to U trapped during the runs.
- c. Includes 35 g overage at Y-12.
- d. Obtained as UF₆·3NaF and in aqueous solutions from purges and washes, respectively.

(Tables 3.1 and 3.2). Product measurements at Y-12 indicated VPP figures to be low by 340 g of uranium. This was attributed primarily to errors in weighing product cylinders because only on one set of product samples did the variation between uranium analyses significantly exceed the accepted limit of error (Sec. 4.0).

4.0 PRODUCT PURITY

The UF₆ product from VPP was of sufficient purity to meet specifications for material designated for reduction to uranium metal. Although several cations were present in excessive amounts for product level UF₆ (to be returned to a gaseous diffusion cascade) the reduction process to green salt reduced all contaminants to acceptable levels (Table 4.1).

Of the cations investigated Cr, Cu, Fe, and Ni most consistently exceeded specifications based on Y-12 analysis, and Al, B, and Ni based on ORNL analysis. Considerable variation was noted in the analysis of samples taken from the same UF₆ cylinder at Y-12 as compared to the ones taken in VPP at ORNL.

Activity and isotopic analyses were acceptable for product level UF₆ (Tables 4.2 and 4.3). Gross gamma and gross beta measurements indicated that enriched uranium was the only radioactive material present in the product. Radiochemical analyses for Cs, Nb, Zr, Ru, TRE, and Te were performed

Table 4.1. UF₆ Product Purity

		Puri- ty*	Impurities, ppm based on U																		
			Al	B	Be	Ca	Cd	Co	Cr	Cu	Fe	Hg	Li	Mn	Mg	Mo	Na	Ni	Si	Ti	V
E-3	ORNL	99.75	12	≥ 37		7			2	22	3					27	< 370	15	**	**	
	Y-12	99.76	< 1.5	< 0.15	< 0.015	37	< 0.15	< 1.5	15	15	< 15		< 0.3	< 0.15	< 3	27		27	< 15		9
E-4	ORNL	99.19	24	≥ 33		3			1	21	12					< 15	< 370	114	**	< 1**	< 1
	Y-12	99.89	< 0.15	< 0.15	< 0.15	< 15	< 0.3	< 0.15	9	30	< 15		< 0.3	< 0.15	< 0.3	22		9	< 15		1.5
E-5	ORNL	99.78	10	≥ 18		2			5	21	8		59			< 15	< 370	37	**	1**	2
	Y-12	99.51	3	< 0.15	< 0.015	30	0.45	< 0.15	119	59	148		< 0.3	1.5	3	30		40	15		11
E-6	ORNL	99.17	4	≥ 4		4			5	16	5					< 15	750	10	**	3**	6
	Y-12	99.73	1.5	< 0.15	< 0.015	45	0.74	1.5	370	126	1,140		< 0.3	22	< 3	37		119	< 15		3
E-1 through E-6 Green Salt		***	< 1.3	< 0.3	< 0.13		< 0.13	< 0.13	< 3	12	20	< 3	< 0.3		< 1.3			16	< 3		< 1.3

$$\text{*Purity} = \frac{\text{wt \% uranium}}{\text{theoretical wt \% uranium}}$$

**Chemical analysis at ORNL indicated excessive Si and Ti, but the results were later shown to be in error. Ti results listed above were from spectrographic analysis.

***Analysis of salt obtained when UF₆ from six ARE runs was reduced to UF₄ prior to reduction to uranium metal.

Table 4.2. Activity of UF₆ Product

Run	Activity,* cpm/mg U	
	Gr γ^{**}	Gr β
E-3	8	206
E-4	10	251
E-5	9	219
E-6	9	210
U Background	11	232

*Analyses performed at ORNL on VPP samples.

**Pb absorber.

Table 4.3. Isotopic Concentration of UF₆ Product

Run	Uranium Isotope,* Weight %				
	234	235	236	238	235/238
E-3	1.15	93.09	0.32	5.44	17.11
E-4	1.20	93.01	0.33	5.46	17.03
E-5	1.17	93.19	0.33	5.31	17.55
E-6	1.19	93.29	0.27	5.25	17.77
Feed Salt**	1.18	92.96	0.32	5.54	16.78

*Analyses performed at Y-12 on VPP samples.

**Average of analyses from Runs E-1, E-3, and E-6.

and only Te γ was detected. However, the concentration of Te observed (~ 2 cpm/mg U) was inconsistent with established data on uranium background (11 cpm/mg U) and the gross gamma count (8-10 cpm/mg U) of the samples. At these low levels of activity, analytical results--which are based on a chemical separation plus a counting procedure--are subject to large relative errors.

Isotopic analyses of product indicated it to be somewhat enriched as compared to the feed, but here again, the variation was considered within the accuracy of sampling and analysis.

5.0 DECONTAMINATION FACTORS

Decontamination was similar to that obtained in the first two ARE runs(¹) in which complete decontamination of the UF₆ product from radioactive fission products was attained. Because calculations based on complete decontamination would yield an infinite decontamination factor, an activity of 1 cpm/mg U was assigned to the product. Thus a fission product gross gamma decontamination factor of $\geq 9 \times 10^4$ was demonstrated during re-

processing of the ARE fuel and apparently was limited only by the low activity of the salt. Decontamination factors, itemized as to individual runs and significant fission products, are listed in Table 5.1.

The fluorination step provided a gross gamma decontamination factor of 500 and a TRE β decontamination factor of 2×10^4 . Fission products which escaped the fluorinator were sorbed on NaF beds (CRP trap, two absorbers, chemical traps) and/or deposited in the lines. Absence of fission product activity in the caustic solution used to scrub the off-gas, and on the filter in the off-gas line, indicated that no significant amount passed through the system. About 80% of the gross gamma activity downstream from the fluorinator was due to Cs-137, and 10-15% of the gross beta in that region was attributed to total rare earths.

The first absorber was a factor of 10 more effective than the CRP trap in removing Cs-137. However, in any evaluation of the above data the low fission product activity in the feed salt (~ 2 curies per batch) and the fission product spectrum for long cooled material must be considered.

6.0 DESCRIPTION OF PROCESS

Fused fluoride salt ($\text{NaF-ZrF}_4\text{-UF}_4$) contained in the ARE dump tank was melted and drained to a hold tank. Salt batches of ~ 170 kg (~ 10 kg U) were pressure transferred to the fluorinator for each run. Fluorine was added to the melt in the fluorinator (maintained at $\sim 600^\circ\text{C}$) at a rate of ~ 15 slm to oxidize the UF_4 to volatile UF_6 . During constant F_2 addition to the melt the fluorination reaction* exhibited three distinct phases: (1) no gas evolution, (2) UF_6 evolution, and (3) F_2 evolution. Although volatilization of UF_6 was $\sim 99\%$ complete at the start of step (3), excess fluorine was added to reduce the final U concentration in the salt to < 25 ppm. Waste salt was pressure transferred to a waste container and taken to a burial ground for radioactive wastes. Volatile fission product fluorides, volatile corrosion product fluorides, and some nonvolatile fluorides (by entrainment) left the fluorinator with the UF_6 .

The UF_6 was separated from these other fluorides in the gas stream by sorption** and desorption in fixed beds of NaF pellets. The gas stream from the fluorinator was routed through the CRP trap (at 400°C), both absorbers (at 70°C), a chemical trap (NaF at room temperature), and a caustic spray tower. Most of the entrained fluorides and some of the volatile fluorides were sorbed (or filtered) in the CRP trap. The UF_6 , plus essentially all the remaining fluorides in the gas stream, were sorbed in the absorbers. The chemical trap served as a safety feature in the event that

**The reaction vessels are called absorbers even though uncertainty exists as to the exact type of sorption reaction that occurs.

Table 5.1. Fission Product Decontamination Factors in ARE Reprocessing

Run	Over-all			Fluorination				CRP Trap				First Absorber***				2nd Absorber***		
	Gr γ	Gr β	TRE β	Gr γ	Gr β	Cs γ **	TRE β	Gr γ	Gr β	Cs γ	TRE β	Gr γ	Gr β	Cs γ	TRE β	Gr γ	Gr β	Cs γ
E-3	$\geq 9.4 \times 10^4$	$\geq 2.3 \times 10^5$	$\geq 1.8 \times 10^5$	580	3.8×10^3	~ 700	3.7×10^4	1.7	1.6	1.5	2.1							
E-4	$\geq 8.4 \times 10^4$	$\geq 2.4 \times 10^5$	$\geq 1.6 \times 10^5$	430	2.9×10^3	~ 500	1.5×10^4	2.1	2.1	1.9	2.5							
E-5	$\geq 1.1 \times 10^5$	$\geq 2.8 \times 10^5$	$\geq 1.5 \times 10^5$	660	4.2×10^3	~ 800	1.5×10^4	1.8	1.7	1.6	2.2	16	12	18	> 4	> 5	> 3	> 4
E-6*	$\geq 9.8 \times 10^4$	$\geq 2.5 \times 10^5$	$\geq 1.6 \times 10^5$	550	3.7×10^3	~ 700	1.5×10^4	1.9	1.7	1.6	2.5							
ARE (Avg)	$\geq 9 \times 10^4$	$\geq 2 \times 10^5$	$\geq 1.8 \times 10^5$	500	3.6×10^3	~ 700	2×10^4	1.8	1.8	1.6	2.1	16	12	18	> 4	> 5	> 3	> 4

*Feed was not analyzed but was assumed to have an average analysis.

**Gr γ in the feed was assumed to be Cs γ .

***Activity in absorbers assumed to be evenly divided among runs.

any UF_6 or radioactive fluorides should reach this point. The remaining gas was scrubbed with 5-10% KOH in a spray tower to neutralize the fluorine and then sent to the off-gas stack. Since the UF_6 -NaF sorption reaction* is forced to the left by increasing temperature, the UF_6 passes through the CRP trap (400°C) and reacts with the absorber bed (70°C). The reaction is exothermic, and external cooling (compressed air) is necessary to control the absorber bed temperature.

After the sorption reaction was completed, the UF_6 was desorbed from the NaF, thereby effecting a separation from other sorbed fluorides which have a different temperature dependence. Desorption was accomplished by heating both absorber beds simultaneously to 400°C in a fluorine sweep gas (~ 8 slm). The oxidizing atmosphere tends to decrease the rate of formation of nonvolatile UF_5 in the absorbers. After leaving the absorbers the UF_6 was condensed in two cold traps. The bulk of the UF_6 was condensed in the first trap, operated at -40°C , and any residual UF_6 vapors were condensed in the second trap, at -55°C . The off-gas was then routed through a chemical trap and a caustic scrubber as in the previous step.

The UF_6 was then liquefied and drained to standard shipping containers by the following procedure. The cold traps and connecting lines were evacuated to < 0.5 mm Hg abs. The system was heated to $> 64^\circ\text{C}$ (UF_6 triple point) thereby liquefying the UF_6 in an atmosphere of UF_6 vapor. This liquid was drained to a product receiver. The product receiver was then cooled to 0°C to effect a thermal transfer of UF_6 vapors. After the thermal transfer appeared complete (indicated by constant product weight), the cylinder valves were closed and the cold traps were cooled to normal operating temperatures. Thus, any UF_6 vapors not transferred were re-condensed in the cold traps and combined with a subsequent run.

The chemistry of the process is described in references 2, 3. Details of pilot plant construction are given in reference 4, and Figures 6.1 and 6.2 illustrate the pilot plant flowsheet.

7.0 PROCESS MODIFICATIONS, LEAK TEST SYSTEM

Prior to resuming the ARE recovery program the entire system had to be proven gas-tight. In previous operations only those valves and fittings directly connected to UF_6 lines were considered critical, but the experience in Run E-2(1) indicated that even those remotely associated with UF_6 lines were possible sources of UF_6 leakage.

Valves and fittings required manual checking. Two technical personnel, one tester and one observer, performed each leak test. Three tests were used in order of increasing sensitivity--(1) soap bubble, (2) flame discoloration by Freon, and (3) fluorine-KI, starch solution reaction. Test (1) involved filling the system with nitrogen to a pressure of 30 psig and then covering the fitting being tested with a soap film. Large leaks would be indicated by the appearance and growth of a soap bubble. Test (2) was

Note: All FV-500 series
are furnaces or heaters

FV- No.	Service
120	First Absorber
121	Second Absorber
124	Chemical Trap
126	Product Receiver
220	5" Cold Trap
222	6" Cold Trap

Fig. 6.2

VOLATILITY PILOT PLANT CELL 2 FLOWSHEET

somewhat more sensitive and consequently more complex. The system was filled with Freon to a pressure of 15-30 psig and then increased to 30 psig with the addition of nitrogen. The flame from a halide leak detector torch was applied to the area in question and a yellow or blue discoloration of the flame indicated leakage of Freon. Test (3) was the most sensitive and therefore used as a final check. The system was filled with fluorine to a pressure of ~ 5 psig, which was then increased to ~ 20 psig with nitrogen. The fitting being tested was cleaned and wrapped with a piece of Kleenex. KI-starch solution was applied to the Kleenex, and the wet paper was pressed against the fitting. After ~ 15 seconds the paper was removed and examined. The presence of a black or brown stain indicated fluorine leakage.

All fittings were identified with a numbered aluminum tag. Then each was leak tested and repaired, if necessary. After this, whenever a fitting was disconnected for any reason, a red tag was attached to it, and a corresponding white tag was filed. Before operations were resumed, all red-tagged fittings had to be repaired.

An HRT type flange leak test assembly was installed to maintain a constant check on the leak-tightness of the flanges on the CRP trap and both absorbers. This involved constant pressure indication of a positive nitrogen pressure of 30 psig placed on the O-ring gaskets of the flange. The system was so arranged that all flanges would be simultaneously checked for total leakage and any leak would be nitrogen into the system. In the event of a leak, individual O-rings could be isolated to locate the leak.

All fittings and flanges were leak tested both at operating temperatures and after being subjected to thermal cycling.

8.0 FEED SALT

8.1 History

The fused fluoride salt ($\text{NaF-ZrF}_4\text{-UF}_4$) feed to the process had previously been used as nuclear fuel in the Aircraft Reactor Experiment (2) in November, 1954. The uranium was fully enriched and had been subjected to 0.006% burnup (96 Mw hours). In the process of removal from the reactor the fuel was diluted with approximately an equal amount of barren salt used to flush out the reactor.

Table 8.1. Physical Constants*

Nominal Composition:	50 mole % NaF, 47 mole % ZrF_4 , 3 mole % UF_4
Density at 600°C:	3.30 g/cc
Melting Point:	520°C $\pm$ 20°C

*After barren salt dilution.

8.2 Composition

Chemical, radiochemical, and isotopic analyses were performed on samples of salt taken from the fluorinator prior to fluorination. The results of these analyses are listed in Table 8.2.

Table 8.2. Feed Salt Analyses

A. Chemical, weight %										
Run	U	Na	Zr	F	Cr	Ni	Fe	Si	Ti	Mo
E-3	5.71	10.82	45.60	37.85	0.0406	0.138	0.0509	0.0030	0.0971	< 0.001
E-4	6.04	10.69	39.3	38.85	0.0458	0.0779	0.0590	0.0015	0.0251	< 0.001
E-5	5.83	11.11		38.85	0.0382	0.0773	0.0518		0.0132	< 0.003
E-6	4.86	10.71	40.2	35.05	0.0311	0.0840	0.0550	0.0026	0.0159	< 0.005

B. Radiochemical, cpm/mg U							
Run	Gr γ	Gr β	TRE β	Zr γ	Nb γ	Ru γ	Te γ
E-3	9.1×10^4	2.3×10^5	1.8×10^5	< 89	< 248	< 195	
E-4	8.4×10^4	2.4×10^5	1.6×10^5	< 63	< 83	< 121	< 25
E-5	1.1×10^5	2.8×10^5	1.5×10^5	< 38	< 60	< 140	< 22

C. Isotopic, weight %					
Run	U-234	U-235	U-236	U-238	235/238
E-3	1.15	92.84	0.30	5.71	16.26
E-6	1.22	92.99	0.33	5.46	17.03

Prior to analysis the salt had been stored in a type 347 stainless steel vessel (FV-114) at $\sim 560^\circ\text{C}$ for periods of 1970 hr (E-3) to 2190 hr (E-6). Also, $\sim 10\%$ of the melt at the time of sampling represented a heel from the previous run (Sec. 9.1). The fission product spectrum was representative of long cooled material except for ruthenium gamma activity. This was lower than would be expected from nuclear calculations and was apparently due to "plating out" in the reactor. (6)

9.0 FLUORINATION

The fluorination operation in the pilot plant consisted of feed salt being transferred to the fluorinator, where fluorine was then added to the melt to volatilize uranium as UF_6 . The resulting waste salt was transferred to a waste receiver and transported to a disposal area for radioactive wastes.

9.1 Plant Operation

The transfer of feed salt from the hold tank to the fluorinator proceeded smoothly in all runs. Transfer was initiated with a pressure of 3-4 psig and could be controlled adequately. The quantity of salt per batch was somewhat larger than the minimum requirements of 39 liters of salt and ~ 10 kg uranium in order to reduce the number of runs required to reprocess the ARE salt (Table 9.1). After Run E-6, salt in the upper leg of the freeze valve (FV-104) was blown back into the hold tank. This line was then cooled and the cross-over pipe was heated so that any residual salt could drain by gravity to the fluorinator. Barren salt (50-50 mole % NaF-ZrF_4)

Table 9.1. Feed Salt Transfer

Run	Heel	Charge	Total Feed in Fluorinator		
	from Previous Run, kg	from Hold Tank, kg	Weight, kg	Volume, liters	Uranium, g
E-3	19.8	172.9	192.7	57	10,868
E-4	13.9	173.1	187.0	56	11,294
E-5	11.6	162.1	173.7	52	10,127
E-6	11.6	133.2	171.8	52	8,349
E-6 (barren salt flush)		27.0			

was transferred from a portable furnace arrangement to the hold tank and then drained to the fluorinator in order to flush all uranium bearing salt from the line.

Fluorination of the four salt batches proceeded without incident. Minor difficulties with salt plugs were encountered in Run E-3 concerning the liquid level instrument probes. Salt was drilled out of the lines and the instrument operated adequately for the remaining runs.

Considerable difficulty was encountered with the waste salt transfer operation. A cold spot ($< 450^{\circ}\text{C}$) at the waste nozzle caused salt to freeze and plug the line at that point. These plugs had to be drilled out in Runs E-4 and E-6. A Calrod heater, wrapped around the waste nozzle to supplement the autoresistance heating in this area, did not completely remedy the situation. Splatter of salt increased the β - γ background around the waste receiver and damaged instrumentation in that area. In Runs E-3 and E-4 several kilograms of salt spilled onto the cell floor causing radiation readings up to 7 r/hr. Poor venting of the waste can while filling apparently contributed to splattering and spillage of salt. In subsequent runs extra holes were drilled in the top of the waste receiver to facilitate venting. In Run E-4 spillage was attributed to an uneven liquid stream flowing from the nozzle. The nitrogen purge rate was increased to remedy this but a siphon developed during the transfer. Consequently, the transfer could not be controlled and the seal in the freeze valve was blown. Prior to Run E-5 barren salt had to be added from a portable furnace arrangement to provide a seal in the freeze valve.

Waste salt transfer operational data are listed in Table 9.2.

9.2 Fluorine Utilization

Fluorine requirements for UF_6 breakthrough and for fluorine breakthrough were about the same and 10-15% less, respectively, than that observed in previous runs⁽¹⁾ (Table 9.3). Total fluorine required for fluorination also was 10-15% less than in previous runs but this was probably more a function of the method of VPP operation than of process chemistry. The fluorine used per run was greater than the amount required to reduce uranium concentration in the salt to < 20 ppm because of the sampling

Table 9.2. Process Data for Waste Salt Transfer

Run No.	Salt Transferred, kg	Heel Left In FV-100, kg	Pressure, psig		Temperature, °C					
			To Initiate Transfer	Max in FV-100	Freeze Valve			Molten Salt Pipes		
					Max	Min	Avg	Max	Min	Avg
E-3	176.4	13.9	4.0	4.4	695	630	662	730	640	685
E-4	170.2	11.6	4.9	6.8	695	625	660	750	645	697
E-5	148.5	11.6	4.3	6.6	710	615	664	795	630	712
E-6	151.9	8.1	4.3	6.3	690	575	632	740	600	670

Table 9.3. Fluorine Utilization

Run	Avg F ₂ Flow Rate, slm	Breakthrough		Total F ₂		U Conc. In Waste Salt, ppm	U Volatilized, % Of Initial
		UF ₆ Moles F ₂ /U	F ₂ Moles F ₂ /U	Required,* Moles F ₂ /U	Actual,** Moles F ₂ /U		
E-3	13.4	0.70	1.5	2.4	3.2	3	99.99
E-4	13.7	0.71	1.6	2.3	3.2	16	99.97
E-5	13.6	0.66	1.8	2.4	3.7	4	99.99
E-6	13.3	0.73	1.9	2.9	3.9	5	99.99

*Necessary to reduce uranium concentration in melt to < 20 ppm.

**Actual fluorine used in the run.

procedure. Initial sampling occurred after ~ 3 mole ratios of F₂/U, with subsequent sampling after one hour intervals (~ 0.8 F₂/U). Only in Run E-6 did the uranium concentration (26 ppm) of the initial samples exceed 20 ppm and this was not considered significant. The salt was fluorinated an extra hour while analytical personnel analyzed a sample.

9.3 CRP Trap Performance

The CRP trap was designed to remove certain fluorides--principally ZrF₄ and CrF₅--from the fluorinator exit gas during fluorination by means of sorption and/or filtration in a bed of NaF pellets. In addition, some decontamination was effected by the NaF bed.

9.3.1 Operation

Operation of the vessel was essentially trouble-free during the four runs. This represented the longest period of pilot plant operation during which no interruption in run procedure was necessary because of malfunction of the CRP trap.

Thermocouples were installed inside the NaF bed at 5-in. intervals along the vertical axis and along a vertical line at the periphery. These indicated that (1) the center of the bed was 5-20°C hotter than the outside edge at the same cross section, (2) the bottom of the bed was 75-120°C hotter than the top, and (3) the bed temperature increased ~ 50°C during UF₆ volatilization (Table 9.4). The initial bed temperature (bottom)

Table 9.4. Operating Temperatures in CRP Trap

Run	Level of Bed Cross Section	Temperature, °C			
		Center		Outside Edge	
		Initial	After UF ₆ Volatilization	Initial	After UF ₆ Volatilization
E-3	Bottom	465	500	460	510
	Top	360	410	345	390
E-4	Bottom	460	515	460	515
	Top	360	445	345	495
E-5	Bottom	425	485	420	440
	Top	345	385	320	375
E-6	Bottom	415	460	410	450
	Top	340	375	320	365

was gradually decreased from 465°C in Run E-3 to 415°C in Run E-6 with no apparent effect on operating characteristics or performance. In Run E-4 the temperature rise of the outside edge of the bed was considerably greater than the center during UF₆ volatilization, thus indicating that gas flow was restricted through the center and had channeled along the outside edge. When the bed was changed after Run E-4, pellets in the central region were found to be agglomerated.

The NaF bed was removed, and fresh pellets were charged after each run. When beds were removed some agglomeration of pellets was observed near the gas inlet, but in general, this could be broken up with little effort. The bottom one-third (gas inlet) of the bed contained a high concentration (> 80%) of orange-tan-brown pellets. The concentration of colored pellets decreased to ~ 30% in the middle third of the bed and to < 2% in the top one-third. A few scattered yellow and green pellets were noted in the top section of the bed. In general, yellow denoted the presence of uranium, and orange-tan-brown-green pellets resulted from successive stages of hydrolysis of CrF₅. This hydrolysis took place when the pellets were exposed to air during sampling.

9.3.2 Retention of U, Cr, Zr, Si, and Ti

Data obtained from analyses of the NaF beds indicated that (1) uranium holdup was sufficiently small except in Run E-5 and (2) chromium removal from the gas stream was 20-30% effective (Table 9.5). Uranium holdup in

Table 9.5. Retention of Materials by CRP Trap

Run	U		Cr		Zr, grams	Si, grams	Ti, grams
	grams	% of Total	grams	% of Total			
E-3	11	0.10	17	26	5.8	30	4.3
E-4	13	0.12	18	22	6.1	33	10
E-5	194	1.92	20	33	< 1	31	3.5
E-6	3	0.04	10	23	3.3	25	3.9

Run E-5 was excessive (194 g) for no apparent reason. Interpretation of Si data was limited by the presence of Na_2SiF_6 in the NaF pellets. Ti investigation was prompted by initial high analysis in the product (later shown to be in error).

Distribution profiles of U, Cr, Zr, and Ti caught in the CRP trap are shown in Fig. 9.1. Only chromium showed a consistent trend--an almost linear decrease in concentration over the first four-tenths to eight-tenths of the bed. In Run E-4 Cr, U, and Ti concentrated in the center section of the bed, probably because of channeling in this run (Sec. 9.3.1). Si data were not plotted because of considerable scatter.

9.3.3 Retention of Fission Products

The CRP trap, although primarily intended to be a trap for chemical contamination, also removed certain fission product fluorides from the gas leaving the fluorinator during fluorination. Radiochemical data from NaF bed samples indicated that 40-50% of the Gr γ activity and ~ 60% of the TRE β activity leaving the fluorinator were caught in the CRP trap. About 70% of this gamma activity was due to Cs-137, the principal long-lived gamma emitter in the feed salt. Since CsF boils at 1250°C, its presence in the CRP trap was probably due to entrainment.

Data from individual runs are listed in Table 9.6 and the distribution

Table 9.6. Fission Product Activity in the CRP Trap*

Run	Gr γ cpm/mg U	Gr β cpm/mg U	TRE β cpm/mg U	Cs γ cpm/mg U	Nb γ cpm/mg U
E-3	60	22		42	8.8
E-4	102	44	6.4	80	11
E-5	72	26	5.4	49	15
E-6	83	28	6.6	55	< 17

*Ru γ , Zr γ , and Te γ were below limit of detection.

of fission product activity is shown in Fig. 9.2. Beta-gamma activity on the bed in Run E-4 exceeded the other runs by 50-70%. This was coincident with high retention of Cr, U, and Ti, and in both cases the materials were

Fig. 9.1. Distribution of Fluorides Retained in the CRP Trap

Legend

U —●—●—
 Cr —▲—▲—
 Zr —○—○—
 Ti —▽—▽—

Fig. 9.2. Distribution of Fission Product Activity in the CRP Trap

concentrated in the middle of the bed (Sec. 9.3.2). Niobium activity was generally reduced to a minimum value ($\sim 10^4$ cpm/g NaF) in the first six-tenths of the bed, possibly because of a sorption reaction with NaF. Cesium showed a reverse behavior--activity in the bed increasing at the top--possibly because of lower temperature in that region. TRE β concentration was essentially constant over the length of the bed.

10.0 SORPTION OF UF_6

UF_6 , mixed with gaseous fluorination products, underwent additional decontamination from volatile fission product fluorides by sorption-desorption* in fixed beds of 1/8-in. NaF pellets. These beds (~ 27 kg of NaF each) were contained in two absorber vessels arranged in series. Each vessel also contained ~ 18 kg of nickel shot which served to disperse the gas prior to contact with NaF.

10.1 Plant Operation

From an operations standpoint, sorption was considered an integral part of the fluorination operation, and therefore any difficulties were included in Sec. 9.1. The bed in each vessel was used for four sorption-desorption cycles (4 runs) and then sampled.

10.2 Absorber Performance

10.2.1 Uranium Loss

No direct measurement was made of uranium losses during the sorption operation. However, a total of 75 g of uranium trapped in the main chemical trap (FV-124) during the four runs indicated that the loss was $< 0.2\%$ of the feed. This absorption loss (UF_6 that passes through the absorber beds without being sorbed) was not considered a process loss because of subsequent recovery of the uranium by aqueous reprocessing methods.

10.2.2 Thermal Characteristics

Control of NaF bed temperatures was necessary to prevent (a) excessive UF_6 losses to the exit gas and (b) deposition of solid UF_6 at the absorber gas inlet (with resulting flow restriction). Temperatures of the absorber bed during sorption of UF_6 are given in Table 10.1 and Fig. 15.1 through 15.4. The exothermic sorption reaction can be followed on these curves (in the absence of an external heat source). Compressed air, directed into the annular space between the absorber and its furnace, was used to limit the temperature rise resulting from the heat of reaction.

Maximum bed temperatures of 160 to 222°C were a function of cooling air operation and UF_6 sorption. Therefore, assuming that cooling air operation was essentially the same, temperature rise was a reasonably quantitative

Table 10.1. First Absorber Bed Temperatures during UF_6 Sorption

Run	Maximum Temperature, °C			Temperature Rise, °C		
	Bottom of Bed	Middle of Bed	Top of Bed	Bottom of Bed	Middle of Bed	Top of Bed
E-3	165	222	115	88	147	53
E-4	131	170	165	61	100	110
E-5	122	160	158	48	88	93
E-6	133	169	120	58	93	52

indication of UF_6 in various portions of a bed. The preferred region for sorption apparently moved up the bed after Run E-3. (Results from Run E-6 were not comparable because the feed contained ~ 20% less uranium). However, in no run did UF_6 break through the first bed and reach the second bed during the fluorination-sorption operation.

11.0 DESORPTION OF UF_6

UF_6 was desorbed from the absorber beds to recover uranium and to provide additional decontamination from those fission product fluorides which also had sorbed on NaF. Desorption was accomplished by heating the absorber beds to ~ 400°C in an atmosphere of fluorine sweep gas. The desorbed UF_6 was then routed through two cold traps in series, where it condensed as a solid.

11.1 Plant Operation

No operational difficulties were encountered during desorption and cold trapping of UF_6 . After Run E-6 the absorbers were disassembled, and the NaF beds were sampled in sections. (7) Process data recorded during the runs are listed in Table 11.1.

Table 11.1. Process Data for Desorption and Cold Trapping Operation

Run	F ₂ Flow Rate, slm	Temperature, °C						
		First Absorber		Second Absorber		First Cold Trap	Second Cold Trap	Scrubber Solution
		Wall	Bed	Wall	Bed			
E-3	8.9	115-450	90-410	80-425	80-410	-46	-53	30-43
E-4	8.9	80-430	80-405	40-430	45-410	-47	-53	34-47
E-5	8.9	50-425	55-400	65-425	70-405	-47	-57	30-42
E-6	8.9	25-425	35-410	55-425	60-410	-47	-55	28-44

11.2 Absorber Performance

11.2.1 Thermal Characteristics

Temperatures of the vessel wall, furnace, and at three points along the bed axis were recorded during the runs. These data are included in the

Appendix as Fig. 15.5 through 15.8. The data were typical of those obtained in previous runs, ⁽¹⁾ showing (1) a plateau at $\sim 310^{\circ}\text{C}$ representing the period of highest rate of UF_6 desorption and (2) a period of ~ 9.5 hr required for complete desorption. Since no uranium reached the second absorber, temperature profiles of that vessel were not included in this report. The rate of temperature rise in the second absorber bed was somewhat greater than in the first absorber, e. g., all thermocouples in the bed reached 390°C about two hours quicker. This was attributed to the fact that no $\text{UF}_6 \cdot 3\text{NaF}$ was present to absorb heat. Also, the plateau indicating UF_6 desorption was absent from temperature profiles of the second absorber bed. Otherwise, temperature profiles of the two absorber beds were essentially the same.

11.2.2 Retention of Uranium and Chromium

Uranium retention (probably as a UF_5 complex with NaF) ⁽²⁾ on the beds represented 0.07% and 0.10% of the total feed for the first and second absorbers, respectively (Table 11.2). However, uranium in the second absorber

Table 11.2. Retention of Materials in Absorber Beds

Absorber	Uranium		Chromium, g	Others
	Weight, g	% of Feed		
First (FV-120)	28	0.07	14	Spot checks for Zr indicated no excessive amounts present
Second (FV-121)	39	0.10	-	

could be attributed not only to desorption but also to a flow reversal during purging of process lines after Run E-6 (Sec. 15.3). Thus, only 22 g of uranium (0.05% of feed) was considered representative of the desorption operation (Fig. 11.1). A uranium concentration profile in the first absorber bed could not be established because of wide scatter of data.

Chromium data throughout the first absorber bed were erratic, possibly because of the very low concentrations in samples. Hence, these analyses were not requested for second absorber bed samples.

11.2.3 Retention of Fission Products

The first absorber effectively removed fission product activity from the gas stream. Gross β - γ activity in the second absorber was a factor of ~ 15 less than in the first absorber, and no significant amount of activity was found downstream from the second absorber. Greater than 90% of the Gr- γ was represented by Cs-137 and $\sim 12\%$ of the Gr β was total rare earths (Table 11.3).

About 75% of the Gr β - γ activity in the first absorber was in the first one-third of the bed past the gas inlet (Fig. 11.2). The Gr γ activity concentration, represented primarily by Cs-137, decreased exponentially along

Fig. 11.1. Distribution of Uranium Retained in the Absorbers

Fig. 11.2. Distribution of Fission Product Activity in the First Absorber

Fig. 11.3. Distribution of Fission Product Activity in the Second Absorber

Table 11.3. Fission Product Retention Absorber Beds

Vessel	Gr γ cpm/mg U	Gr β cpm/mg U	TRE β cpm/mg U	Cs γ cpm/mg U	Nb γ cpm/mg U
1st Absorber (FV-120)	89	36	4.4	83	≥ 1
2nd Absorber (FV-121)	5.2	2.8	-	4.3	-

the direction of gas flow through the bed. However, in contrast to its behavior in the CRP trap, Nb-95 concentrated at the top of the bed. Nb γ was detected in only the top two samples (top 15% of bed).

Activity distribution in the second absorber was considerably different, with the top 25% of the bed containing > 50% of the gross β - γ activity (Fig. 11.3). Total activity was small, and the only specific activity detected was Cs-137. It is conceivable that the flow reversal during purging caused fission products to deposit in the top section of the bed. This would indicate that some activity passed through the second absorber bed during the runs and remained in the system.

12.0 PRODUCT TRANSFER

Solid UF₆ contained in the cold traps was liquefied by the addition of heat and the resulting UF₆ vapor pressure.* After the liquid was drained to the product cylinder, residual UF₆ vapors were recovered by thermal transfer.

12.1 Plant Operation

No major difficulties occurred during the transfer of UF₆ product from the cold traps to the product receiver. In Run E-3, however, transfer was delayed by a suspected line plug which necessitated extensive pressure testing. During the testing procedure, a new product cylinder was installed and the final 410 g of UF₆ was collected in this cylinder. A pressure build-up of 5 psig in the system apparently was caused by nitrogen inleakage through a valve.

As in previous runs the product sampling operation was troublesome. The sampling manifold became plugged on several occasions, and small quantities of UF₆ were lost (< 5 g total). Constant air monitors indicated excessive air contamination at those times (Sec. 13.0).

Pertinent data on run conditions recorded during the product transfer operation are presented in the Appendix in Fig. 15.9 through 15.12.

12.2 Recovery

Product from each run was collected in a standard shipping cylinder (25 kg UF₆ capacity), weighed, and sampled (Table 12.1). The samples were

*UF₆ triple point at 64°C and 22.4 psia.

analyzed by chemical, radiochemical, spectrochemical, and mass spectrographic techniques (Sec. 4.0).

Table 12.1. Product Recovery

Run	UF ₆ , g	U Analysis, g/g	Uranium	
			Weight, g	% of Feed
E-3	14,360	0.6718	10,037**	92.35
	410*	0.6613*		
E-4	16,910	0.6613	11,183	99.02
E-5	14,390	0.6720	9,670	95.49
E-6	12,750	0.6679	8,516	102.0

*Run E-3 product from E-4 product cylinder which was installed before completion of the E-3 product transfer.

**Includes 390 g from E-4 product cylinder.

The product cylinders were shipped to Y-12, where the UF₆ was removed. After their return, these cylinders were washed out to remove any residue. Analyses of the wash solutions indicated that 105 g of uranium had remained in the cylinders (Table 12.2). Each cylinder was washed three times, first

Table 12.2. Clean-out of "Empty" Product Cylinders

Run	Cylinder No.	Uranium, g
E-1	12	15
E-2	13	24
E-3	15	14
E-4	14	36
E-5	1	13
E-6	2	3
TOTAL		105

with ~ 80 liters of 5% (NH₄)₂CO₃ and then two washes of ~ 80 liters of water each. Greater than 80% of the uranium was removed with the first wash (conc. ~ 0.2 g/l), and < 0.1 g of uranium was present in the third wash.

12.3 Chemical Traps

Three chemical traps, fixed beds of NaF pellets at room temperature, were in the system primarily to sorb uranium from off-gas streams. During Runs E-3 through E-6 these traps retained 248 g of uranium, and an additional 335 g was collected during the post E-6 purging (Table 12.3). This uranium was not considered a process loss because subsequent recovery was made by dissolution of the NaF, followed by aqueous reprocessing.

Table 12.3. Uranium Retained by Chemical Traps

Vessel	Bed Service Life	Uranium	
		Weight, g	% of Feed
FV-122	Runs E-3, 4	147	0.66
	Runs E-5, 6	19	0.10
FV-124	Runs E-3, 4	14	0.06
	Runs E-5, 6	61	0.33
	Post E-6 purge (1)	286	0.82
	Post E-6 purge (2)	49	
FV-158	Runs E-1 through E-6	7*	
TOTAL		583	1.43

*About 50 g of total U was in the trap, but α pulse analysis indicated that only 7 g was enriched U and therefore attributable to "E" runs.

The main chemical trap (FV-124) and the vacuum line chemical trap (FV-122) were used for two consecutive runs before the beds were recharged. Vessel FV-124 was also used to trap uranium during clean-out of the system, (post E-6 purging). The bed in the chemical trap for the heated duct system (FV-158) had been in service during the entire "E" series of runs.

13.0 PERSONNEL EXPOSURE

Biological radiation hazards to personnel were (1) α activity from uranium and (2) β - γ activity from fission products in the feed salt.

Constant air monitors in Cell I and Cell II indicated only one case of excessive air contamination during Runs E-3 through E-6. A leaky flange on the second absorber apparently allowed a small amount of UF_6 (probably < 1 g) to escape during fluorination in Run E-5. A half-hour air sample at that time contained 1.2×10^{-9} $\mu\text{c/cc}$.

The greatest β - γ radiation levels existed around the fluorinator and the waste salt carrier. Waste salt spills in Run E-3 and E-4 resulted in areas of radioactivity up to 7 r/hr, as measured by a "cutie pie." After the spilled salt had been cleaned up, β - γ background was ~ 30 mr/hr in Cell I-A. The dose rate at the outside edge of the fluorinator was ~ 80 mr/hr and the CRP trap had a reading of 30-40 mr/hr. Cell II equipment was checked in place with a "cutie pie" but no significant readings were obtained.

Special film badges were worn by VPP personnel during Runs E-3 through E-6 to permit weekly measurements of exposure. During this two week period,

no overexposures to VPP personnel occurred (Table 13.1). Analysis of urine samples for this period showed that no significant internal exposures occurred.

Table 13.1. VPP Personnel Exposures during Runs E-3 through E-6

Personnel	Dose, mrem			
	<u>Week Ending 3-6-58</u>		<u>Week Ending 3-13-58</u>	
	Average	Range	Average	Range
Operators	57	40-75	36	15-70
Technical	34	0-55	49	0-160

14.0 REFERENCES

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15.0 APPENDIX

15.1 Run Schedule

Table 15.1. Run Schedule

Run No.	Started	Time Started	Ended	Time Ended	Total Time, hours
E-3	2-26-58	0625	2-28-58	1555	46
E-4	3- 1-58	0450	3- 3-58	1200	42
E-5	3- 4-58	2020	3- 6-58	2050	48
E-6	3- 6-58	2145	3- 9-58	2309	73

15.2 Accuracy of Calculations

The various measurements used in material balance calculations were subject to certain limits of accuracy (Table 15.2). Propagation of errors

Table 15.2. Accuracy of Data

Calculation	Measurement	Limit of Uncertainty
Feed	Weight (LR-2)	± 1.8 kg
	U analysis, potentiometric	$\pm 0.5\%$
	U analysis, isotopic	$\pm 0.01\%$
Product	Weight	± 28 g
	U analysis, potentiometric	$\pm 0.2\%$
	U analysis, isotopic	$\pm 0.01\%$
NaF Dissolutions	Volume	± 1.5 liters
	U analysis, fluorimetric or colorimetric	$\pm 5\%$

for a calculation involving more than one measurement was determined by the square root of the sum of the squares technique. These variances represent minimum values because of additional errors, e. g., precision of sampling, for which limits of error have not been established. The difference in uranium content as calculated from duplicate samples was compared to the limit of uncertainty calculations in the material balance. In general, $\pm 10\%$ was estimated to be the limit of uncertainty on all calculations for uranium in NaF dissolutions.

Results from radiochemical analyses were based on counting rates determined through a lead absorber. Counting efficiencies were as follows:

Cs γ 18.5%	Nb γ 21.5%	Zr γ 21.4%
Ru γ 15 %	Te γ 1.2%	TRE β 4.4%

15.3 Equipment Clean-out

Clean-out of the system was limited to nitrogen purges through the piping and the cold traps. It was recognized that this method was inadequate for complete uranium recovery, but water flushing was undesirable because of the impending program of uranium recovery from salt used in criticality experiments. Complete removal of moisture from the system is difficult, and its presence results in problems from the hydrolysis reaction with uranium and increased corrosion.

The entire system was purged with N_2 for about one-half hour at 7 slm and one-half hour at 20 slm. Flow was directed through the chemical trap (FV-124) to collect any UF_6 purged from the system. FV-124 was then disassembled, and the NaF bed was dissolved and sampled. However, during subsequent leak-testing of valves, some white smoke was observed. Since this is indicative, although not necessarily proof, of UF_6 the line was purged with nitrogen for ~ two hours back through the two absorbers. Then FV-124 was reinstalled and the system purged again. This time various flow paths were used (all directed through FV-124) for about two hours each, the flow rate being ~ 21 slm. Thus, the total flow time was 14 hr. The bed from FV-124 was again dissolved and sampled.

Analysis of NaF bed dissolutions indicated that 286 and 49 g of uranium were trapped in the first and second beds, respectively; and an additional 17 g in the second absorber was attributed to purging (Sec. 11.2.2). This material can be classified as the holdup for the product transfer system. Another 50 to 100 g of uranium is estimated to have remained in the system, primarily in the cold traps.

15.4 Uranium Accountability

Uranium material balance figures reported in Sec. 3.0 represent process measurements at VPP. Additional losses were incurred in preparing the various materials for shipment to Y-12, where each was eventually processed to metallic uranium (Table 15.3). These nonprocess losses occurred in

Table 15.3. Uranium Accountability

	Uranium, grams
Product	59,136
Aqueous solutions*	1,516
Analytical waste	118
Total received at Y-12	60,770
Total process recovery (Table 3.2)	<u>60,940</u>
Nonprocess loss	170

*Includes 1,434 g from NaF dissolutions and 82 g from wash of product cylinders.

(a) sampling of UF_6 product cylinders, (b) handling large volumes of aqueous solutions, and (c) discarding of NaF bed dissolutions containing < 5 g of uranium per 55-gal drum which were considered uneconomical for uranium recovery by aqueous reprocessing.

15.5 Process Data

Data recorded during Runs E-3 through E-6 are shown in Fig. 15.1 through 15.12.

The legend for Fig. 15.1 through 15.8 is as follows:

<u>Code</u>	<u>Absorber</u>	<u>Position of Thermocouple</u>
A	First	Axial center of bed, near top (outlet)
B	First	Axial center of bed, near middle
C	First	Axial center of bed, near bottom (inlet)
D	First	Wall of vessel
E	First	Furnace
F	Second	Axial center of bed, near bottom (inlet)
G	Second	Wall of vessel

Fig. 15.1. Temperatures in the Absorbers during Absorption of UF_6 in Run E-3

Fig. 15.2. Temperatures in the Absorbers during Absorption of UF_6 in Run E-4

Fig. 15.3. Temperatures in the Absorbers during Absorption of UF₆ in Run E-5

Fig. 15.4. Temperatures in the Absorbers during Absorption of UF₆ in Run E-6

Fig. 15.5. Temperatures in the First Absorber during Desorption of UF₆ in Run E-3

Fig. 15.6. Temperatures in the First Absorber during Desorption of UF₆ in Run E-4

Note: Delay to remove plug in scrubber inlet.

Fig. 15.7. Temperatures in the First Absorber during Desorption of UF₆ in Run E-5

Fig. 15.8. Temperatures in the First Absorber during Desorption of UF₆ in Run E-6

Fig. 15.9. Product Transfer Conditions in Run E-3

Fig. 15.10. Product Transfer Conditions in Run E-4

Fig. 15.11. Product Transfer Conditions in Run E-5

Fig. 15.12. Product Transfer Conditions in Run E-6

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