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RECOVERY OF URANIUM FROM ALPHA-II CALUTRON

EQUIPMENT

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by

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2

ABSTRACT

This paper describes Recovery Studies made with Alpha-II Calutron equipment in the interval from October 1944 through December 1944 and a study made of the Alpha-I/5 Liner in February, 1945, all at Berkeley. Mention is made of subsequent work done in the Alpha-II plant in the Y-12 area at Oak Ridge, Tennessee in May and June 1945.

It is shown that it is possible to account for $97.5\% \pm 1.2\%$ of the uranium input in a series of twelve runs. The chemical recovery alone averaged 94.5%. The equipment consisted of one Calutron tank, two sources, two liners, and two collectors.

Descriptions of the equipment, the chemistry of the substances involved, and the methods employed for recovery are given in detail. The use of beta-count surveys for establishing the extent of the variation of the equipment and for accounting for a small amount of the total uranium is described, and a complete discussion of uncertainties and errors is included.

RECOVERY OF URANIUM FROM ALPHA II CALUTRON EQUIPMENT

OUTLINE

Abstract

I. RECOVERY STUDIES

1. Introduction
2. Purpose and Limitations
3. Conduct of the Recovery Studies
4. Outline of the Recovery Studies

II. CALUTRON EQUIPMENT

5. Description of the Alpha II Calutron. Accessory Facilities.
6. Operation of the Calutron Equipment.
7. History of the Units used in the Recovery Studies
8. Corrosion of Units during Recovery Operations

III. CHEMISTRY OF THE CALUTRON AND OF RECOVERY FROM THE UNITS.

9. Chemistry of UO_2 Gas.
10. Products of the Arc
11. Reactions of Products from the Source with the Calutron Equipment. The Deposits.
12. Changes in the Deposits upon contact with Ordinary Air
13. Removal of the Deposits.

IV. THE EQUIPMENT USED TO PERFORM THE RECOVERY STUDIES

14. The Equipment used to perform the Recovery Studies

V. RECOVERY OPERATIONS

15. Operations as a Whole (see also section 4)
16. Preliminary Operations
17. Recovery Operations on a Source
18. Recovery Operations on a Liner
19. Recovery Operations on a Collector
20. Recovery Operations on a Calutron Tank
21. Salvage Operations
22. Treatment of Solutions and Sludges

VI. DETERMINATION OF QUANTITIES UNCERTAINTIES AND ERRORS

23. Normal Uranium Input.
24. Normal Uranium in Solutions and Sludges.
25. Normal Uranium in Salvage.
26. Enhanced Uranium. The Product.
27. Impoverished Uranium.
28. Uranium Located but not Gathered.
29. Uranium Lost.
30. Variation of the Uranium Content of the Calutron Equipment

VII. RESULTS OF THE STUDIES

31. Recovery Results.
32. Consequences of the Final Results

33. APPENDIX I. Study of Recovery in Alpha II at Oak Ridge, Tenn. --Y-12.

- 34. APPENDIX II. Recovery of Uranium from a Source Scheduled for Repairs.
- 35. APPENDIX III. The Beta Count.
- 36. BIBLIOGRAPHY

I. Recovery Studies

1. Introduction

A recovery study is actually a study of the amount and nature of the loss encountered in a process. To accomplish the goal, attention must be paid to every detail of the recovery operations, to inspection of the equipment, to investigation of possible sources of loss, and to determination of the uncertainties and errors in measurements. A fairly large amount of accessory information must also be available.

It is unfortunate that the method of study is by necessity indirect, since it is desirable to measure directly quantities which are small, such as one hopes that actual losses will be.

In the Calutron Process of separation of U_{235} , recovery may be divided into two parts: (a) the recovery from the Calutron equipment, which results in a group of solutions and solids, and (b) separation of the uranium from these solutions and solids by chemical means and the preparation of the pure UCl_4 required for recycling the uranium back through the Calutron Process. The research plant at Berkeley, California was not set up to perform the second portion of the recovery study on the scale required for Alpha-II units, and this was therefore omitted.

2. Purpose and Limitations.

The state of affairs in the Alpha-II Calutron plant in the Y-12 area at Oak Ridge in the Fall of 1944 was such that it was not possible to tell where the uranium was disappearing. It was eventually realized that a large portion of the material was on the walls of the Calutron tanks.

However, these tanks were of such construction as to render quantitative removal of the uranium from the walls, roof, floor, and vacuum manifolds a difficult task, and it was decided that liners were to be constructed and tested for use. The purpose of the liners was to lower the amount of material contributed to the walls of the tank and thus held out of the process. The liners were also to be removable from the tank at the end of each run, if desired. It was decided to perform recovery studies with the liners at Berkeley, where the first liners would be constructed, and that recovery studies were to be started in the Alpha-II plant in the Y-12 area as soon as liners were available there.

The purpose of the study at Berkeley may therefore be summarized as follows:

- (1) To test the effectiveness of liners, estimating the amount which escaped the liner and went to the tank walls
- (2) To determine the ease of cleaning the liners, so as to help determine expansion of facilities in the Y-12 recovery plant
- (3) To gather information on total effort, cleaning methods, and chemical methods
- (4) To obtain the information prior to the time it would be gathered at Y-12
- (5) To provide a convenient place for observation of the work by those who were concerned with the overall direction and design of the Calutron process and units
- (6) To provide a sufficiently independent study of the recovery so that incentive might be given to those working under more difficult conditions at the plant
- (7) To train those doing the work in Berkeley so as to be of better service. (See section 33)

used in the Alpha-II plant to the largest extent. The work

meetinghouse designed later, the type being that eventually

II. (November-December 1944). A study of the recovery using a

length.

with the XAH-2 catalyst. The runs were both of reasonable

designed later. The work extended through runs 35 and 36

I. (November 1944). A study of the recovery using a new UCL

The recovery studies may be outlined as follows:

4. Outline of the Recovery Studies.

a beta particle count. (See sections 26 and 27)

estimation of hold-up and losses to equipment was to be made by means of

of the recovered solutions and solids. (See sections 24-26). An

The determination of the uranium found was to be by proper sampling

(See section 2 and section 23)

of the containers and for volatilization of impurities from the UCL.

containers in which the UCL was placed, making a correction for corrosion

that the best method was that of measuring the weight loss of the con-

into a given recovery study be as direct as possible. It was decided

was decided that the determination of the amount of uranium which went

On account of the shortness anticipated for some of the runs, it

5. Conduct of the Recovery Studies.

sub.

next ten were rather short, and the recovery work had to be altered to

made for the purpose of recovery were of proper length, but eight of the

departmental program of the Berkeley research group. The first two runs

chemical plant and the need of tying in the recovery work with the ex-

The two limitations placed on the study were the absence of the

was continued over ten runs in the XAH-2 Calutron at Berkeley, runs 36-45. Runs 36 and 45 were of reasonable length. All the others were short and recovery methods were modified to suit.

III. (February 1946). A test of a liner being considered for use in Track 5 at Y-12 in the Alpha-I plant. The liner was tested over four short runs in the XAH-2 Calutron, runs 54-57.

(See section 7 for the history of the units during the recovery studies.)

II. Calutron Equipment

5. Description of the Alpha-II Calutron. Accessory Facilities.

The various units which constitute a Calutron have received adequate description in other UCRL collected papers. The brief description which follows serves the purpose of designating the units and telling briefly the materials of which they were constructed.

The Calutron Tank. The Calutron tank used in the studies was the XAH-2 unit at Berkeley. This was similar to the tanks used in the Alpha-II plant at Y-12, but differed in the vacuum connections. The tank consisted of a rectangular box placed between the poles of a large magnet. The side walls were about two inches thick and were double, allowing the circulation of hot and cold water through them. The height of the box inside was twelve feet, the depth seven feet and nine inches, and the width twenty four inches. Magnetic shims, shaped somewhat as a section of a parabola ran vertically from near the floor to near the ceiling in the back inside half of the tank. These were bolted to the inside. The thickness of the shims was about two inches at the widest point. The surfaces were rough.

The construction throughout was of non-magnetic manganese steel.

Vacuum manifolds were connected at the upper rear and veered off to

one side so that material could not go in a straight line from the front of the tank into the major portion of the vacuum manifold.

The tank was outfitted with a large liquid air trap. There were also large and small ports intended for observation plates.

The Door and Liner. The front of the Calutron tank consisted of a large manganese steel plate, called the door. There were two large openings, one for the source and the other for the receiver. On the door were mounted either a stainless steel framework bearing several water-cooled copper plates and one or more carbon plates, or a rudimentary liner consisting of water-cooled side plates, end plates, and defining-vane carbon plates, or one of the liners designed for recovery. The recovery liners, whether UCRL, Westinghouse, or Alpha I/5, were similar in design. They consisted of a large hollow container with water-cooled copper sides, about 1/8" thick and properly ribbed for strength. A pumpout opening was located along the periphery and consisted of a copper sheet held about three inches away from the main body of the liner by supporting ribs. The opening was faced with defining-vane copper or graphite sheets, set at appropriate angles. There were also a large number of graphite plates mounted inside which served either as defining vanes or as a protection to the copper. The Westinghouse Alpha-II liner is shown in outline in Figure 1, and photographically in Figure 1a.

The Source. The source of the Alpha-II Calutron consisted of a heavy, intricate, apparatus weighing out 2000 lbs. and consisting of two frameworks, one of which was insulated. The insulated framework was a bronze casting. It contained the two stainless steel charge bottles for the UCl_4 , each of twenty lbs. capacity, with appropriate heaters and shields, connected to a vapor passage lined with nickel and leading to a total of four rectangular boxes about 3.5" x 3.5" x 16" in size. These boxes were

ALPHA II-LINER

Figure 1

10

ALPHA-II LINER

Figure 1a

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the spaces in which the arcs were kept going in UOCl_4 gas. The walls were of graphite. The anticathode was made of vapor and heat resistant material. The cathode was contained in a small box at one end of the arc box and consisted of a $1/8$ " diameter tantalum filament mounted in a cartridge which could be pulled out through a vacuum lock. Also mounted to the insulated bronze frame were several tungsten drain fins placed between the arc boxes.

The uninsulated frame surrounded the insulated one and reached out to the front. It consisted of a heavy bronze casting mounted directly on the manganese steel faceplate. Another casting, rectangular in shape, was mounted in front and formed a window to the front of the arc boxes. Heavy copper inter-arc fins, a set of uninsulated graphite rods, and four insulators bearing a stainless steel framework which supported an insulated set of graphite rods, were all attached to the rectangular frame. The casting which was attached directly to the faceplate supported a number of heavy copper heat shields and electron-drain shields.

The front of the arc boxes was open over an area about a half inch wide and sixteen inches long and faced into a system of graphite rods maintained at proper voltages, to constitute the accelerating system.

The chief materials of construction were bronze, copper, stainless steel, graphite, nickel, molybdenum, tungsten, ceramics, and mica. There was also the manganese-steel faceplate. Silver solder was used throughout for repairs and for attaching water lines, and it gave considerable difficulty in the recovery work. The bronze and copper castings were usually quite porous.

12

The Collector. The collector consisted of a manganese steel face plate from which a fairly large steel plate was supported by posts. The plate served as a mounting for four collectors, corresponding to the four arcs of the source. Each of the collector cages was constructed primarily of sheet brass and cast bronze, and it housed gear and lever mechanisms for proper adjustment of the parts. To these cages heavy graphite pieces were pinned, and behind these a light copper or stainless steel U_{235} collecting-can was set. Copper screens were mounted on the underside of the collector cages in such a position as to collect U_{235} deflected from the graphite piece opposite it.

Accessory Facilities. For proper servicing of the units there was a source repair and assembly department and a collector repair and assembly department. Recovery work was done with the cooperation of both of these departments.

6. Operation of the Calutron Equipment.

The cycle of operations composing a run with the Calutron equipment consisted of installation of the equipment, pumping out the air, running hot water into the side walls, starting the diffusion pumps, turning on heaters in the source, striking the arcs and gradually building up the voltages in the accelerating system, all with the intent of baking water out of the system. This process would require from 12 to 36 hours. When vacuum and arc conditions were satisfactory, cold water was put into the tank sidewalls and reception or experimentation was started. Runs were terminated either by intention, or for repair, or on account of exhaustion of the UCl_4 . The Calutron was then let down to dry air, and the equipment was removed.

13

The only interest one has in the runs, during the operations, is in providing standby recovery service for the rather frequent filament replacements. The statistics for a run are of importance, however, to the person who is responsible for the recovery study, and it is normally desirable to keep a fairly good watch of the operations and experiments.

7. History of the Units Used in the Recovery Studies.

The history of the units involved in Recovery Studies Series I and II is shown in Table 1. The significance of the operations is discussed in sections 15 to 21.

The operations for Study III are not listed in the table. For this study the XAH-2 tank was first washed down and dried, then lined with 0.005" thick copper foil, held on with small brass drive-in screws. The determination of the uranium on the foil was made chemically. The new Alpha 1/5 liner used in the test was washed and scrubbed so as to compare it with the previous liners tested. The source and the collector were not included in the study, since the foil was intended to give a direct answer to the chief question.

Table 1 on next page

14

Table 1

SUMMARY OF THE HISTORY OF UNITS
AND OF RECOVERY OPERATIONS

<u>Key Letter</u>	<u>Recovery Operation or History</u>
A	Steam and water wash without scrubbing
B	Unit disassembled down to soldered connections then rinsed with steam and water a second time
C	Disassembled metal parts given a light scrub
D	Carbon pieces scoured with wire brushes
E	All carbon parts set aside for salvage
F	Carbons ashed for assay
G	Swept up loose material
H	Dry cleaned the surfaces, using a hammer, light anvil, brushes and scrapers
J	Sprayed with 5% ammonium carbonate, scrubbed, removed surface scale from exposed surfaces
K	Complete surface cleanup (See Appendix II)
L	Washed surfaces for one hour with 10% citric acid at 50°C, lightly brushed the surfaces and rinsed with water
S	The tank was thoroughly sandblasted and then vacuum cleaned
beta	An estimate of the remaining uranium was made by means of a beta particle count

Series	XAH-2	Run Length milliamperes hours	Source		Liner		Receiver		Calutron Tank
	Run No.		"Gregory"	"Geofy"	UCRL	Westing- house	"Dorothy"	"Alpha- Gloria"	XAH-2
I	Prior History		ABEJ		New		Clean		S
	33	29970	AJD		AJD, beta		EJ		beta
	34	32700	ADEFJ		AJDEJ, beta		EJ		beta
II	Prior History		K		New			New	S, beta
	36	44500			AEJ, beta		ABGJ, beta	beta	beta, G
	37	16550	AEJ						
	38	9734		ABCDE					
	39	7204	ABCD				G		
	40	9412		ABCD		A	FGH		G
	41	23000		ABCD			FG		
	42	6220							
	43	3450	ABCD			A			G
	44	8300	ABCD						
	45	32700		ABC		ABC	FG		G
	Final Treatment		EJ	EJ beta		EJ, beta	EJ beta	beta	L, beta

8. Corrosion of Units during Recovery Operations.

Recovery of material from units results in an important advantage and an important disadvantage. The extent to which the units themselves deteriorate during a recovery operation due to the action of the solutions may be serious. On the other hand, a thoroughly cleaned unit may be easier to repair and to assemble, and it is likely to bake out in much less time when put into use.

It was found that in the steam and water washes of a source the total amount of copper was 100 to 220 grams, the iron 27 grams, and the nickel 5 grams. Likewise for the liner the copper was 150 to 260 grams, the iron 37 grams, and the nickel 3 grams. Chromium and other metals were not determined. Most of the iron and copper found may be regarded as having been oxidized directly by UCl_4 vapor or by chlorine from the arc, and is not to be ascribed to the action of steam and water.

The washing with 5% ammonium carbonate solution allowed the copper to be oxidized by oxygen of the air; the copper corrosion was large. The corrosion of other metals, except nickel, was probably rather small. The copper corrosion for a source averaged 1230 grams per wash, for a liner 1650 grams, and for a receiver 800 grams per wash.

The corrosion of the Calutron tank was not a serious problem for the tank itself but it would have been for the chemist responsible for salvaging the uranium from the wash, since the iron content was high. Some corrosion of the walls occurs directly from the action of Cl_2 and HCl from the units in operation. An important second factor is the catalytic action of the syrupy UCl_4 solution which forms on the walls upon exposure of the tank to ordinary air. This is oxidized by the

16

air, then reduced by the iron, and the process is repeated as contact with air and iron is repeated. A third factor is the action of the wash solution. The only direct measurement of the corrosion of the tank walls was that obtained on washing down the tank with 10% citric acid at the end of run 45. The tank had been sandblasted prior to run 36 and then nothing had been done with it before the final wash after run 45. The tank was opened to the air nine times, but was protected from gross action by UCl_4 on account of the protection given it by the liner used in the series. The iron found was 1550 grams, the copper 127 grams, and the chloride 298 grams. The presence of the copper indicates the volatilization of $CuCl_2$ from the source and liner. The uranium and copper found in the wash solution account for three-fourths of the chloride, indicating that only about 4% of the iron in the wash solution was brought there as a result of attack by HCl or O_2 .

III. CHEMISTRY OF THE CALUTRON AND OF RECOVERY FROM THE UNITS.

9. The Chemistry of UCl_4 Gas.

Solid UCl_4 as used in the charge containers for the sources at Berkeley was sublimed before being used. Its chief impurities were small amounts of $FeCl_2$, $FeCl_3$, $CrCl_2$, $NiCl_2$, traces of $MoCl_5$, and HCl due to reaction of the solid with water during handling. The $UOCl_2$ contained in the solid was of no consequence in the recovery studies. The iron content averaged 0.3%, the chromium 0.06%, the nickel 0.03%, and the HCl 0.05% $\pm$ 0.01%. Upon being heated in a vacuum to 450°C the HCl and $MoCl_5$ were completely removed, and $FeCl_3$ was partially removed. The chromium and nickel were essentially unchanged. Copper was not present

17

in sufficient amounts in the charge to be considered, but had it been there some CuCl_2 and CuCl would have been lost.

The total HCl loss amounted to four grams per charge bottle.

UCl_4 gas is a fairly strong oxidizing agent. Studies of the reaction of the gas at temperatures ranging from 450° to 500°C (Ref. 9 and 10) show that the gas reacts readily with copper metal to produce both CuCl_2 and CuCl , and that little protection is offered to the metal by the UCl_3 which is left behind. Iron metal is attacked also to form both FeCl_2 and FeCl_3 (or Fe_2Cl_6 as a gas), and the film of UCl_3 is not a good protection for the metal. Nickel was found to be subject to very slight corrosion up to 500°C , due to the non-volatility of NiCl_2 , but found to corrode rapidly at 600°C or above. Chromium was fairly readily attacked but the corrosion of the metal was not rapid. However, in the corrosion of stainless steels by UCl_4 gas, nickel was the chief metal left behind. Both the chromium and iron were removed. Other studies have shown that zinc, lead, and tin undergo extremely rapid corrosion. Silver in the hard solders did not seem to have undergone rapid corrosion, probably due to the low temperature at which most of the hard solder was kept. Graphite was stable toward UCl_4 gas. Ceramics did not react, but slowly acquired a surface coat of reddish or yellow uranium glass if the ceramics were hot enough. The magnesium oxide packing inside Calrods was found to accumulate UO_2 fairly rapidly, probably on account of the slow removal of water from these packings. The UO_2 lowered the electrical resistance of the packing.

The reaction with the stainless steel of the charge bottles was

18

important. It was determined in Recovery Study II that about 6/7ths of the corrosion of the bottles occurred while the bottles were in the Calutron unit, the remainder occurring during the washing. The average corrosion per bottle per run for Studies I and II was 20 grams, with a range from 5 grams to 55 grams.

The overall change in the iron, nickel, and chromium content of the charge bottle and its contents per 1000 grams of UCl_4 vaporized in Studies I and II was determined to be a loss of 17 grams of $FeCl_3$, a gain of 1 gram of $CrCl_2$, and a loss of about 3 grams of $NiCl_2$.

10. Products of the Arc.

The products of the arcs in the source are U^+ , U^{++} , Cl^+ , Cl^- , UCl_x^+ sidebands, O^+ , Cu^+ , neutral Cl , UCl_4 , U , C , Cl_2 , plus small amounts of metal ions such as Ta^+ . The composition of the arc products varies with conditions, but it may be assumed that at least 80% of the UCl_4 entering the arc escapes unchanged. Small amounts of C_2Cl_6 are formed by the reaction of Cl_2 or Cl on graphite somewhere in the source. This material is partly responsible for the odor of a source when it is removed from the Calutron tank.

11. Reactions of Products from the Source with the Calutron Equipment. The Deposits.

The reaction of the products from the source with their environment is difficult to state with assurance. It is best to reduce the matter to what is known concerning the gas phase and the deposits.

The chief gas in the tank, not including the products of the source, during the bakeout period is water. There is also some HCl . The H_2O is gradually eliminated, while HCl is increased. During the reception portion of a run the chief gases present are HCl , H_2 , and CO . Cl_2 apparently does not exist long enough to have more than one

19

collision with a hot metal surface.

The deposits on the source appear to be primarily UCl_4 , UCl_3 , UOCl_2 , C , CuCl , and some basic copper chlorides, together with small amounts of other metallic chlorides and oxides. The deposits on the collector are chiefly U , UC_2 , and UO_2 . The deposits on the liner near the source resemble those of the source and near the collector those of the collector. The periphery of the liner and the side plates in the collector half are covered with a hard black film which is probably a mixture of U , C , and UC_2 .

12. Changes in the Deposits upon contact with Ordinary Air.

It is characteristic of the metallic uranium deposits to react readily with oxygen of the air. Sometimes the uranium films on the receiver begin to burn when the tank is let down to air. The deposits of uranium at any place on the units may be assumed to be covered over at least with a thin film of UO_2 .

Uranium carbide is also somewhat pyrophoric.

Uranium trichloride is gradually oxidized.

All deposits of UCl_4 and UOCl_2 absorb moisture readily and dissolve themselves into thick syrups. These drip from the units.

13. Removal of the Deposits.

Removal of the deposits by abrasion was an obvious and necessary one of the methods used by the recovery group.

Chemical removal of the more adherent deposits required the use of oxidizing agents combined with any reagent which would bring the oxidized substance into solution.

20

Nitric acid was an obvious choice but suffered the disadvantage of reacting rapidly with copper. This acid was used widely on stainless steel units in the Beta plant. It has been used for experimental purposes on collectors in the Alpha plant.

Hydrochloric acid was effective by itself in its action on uranium halides, but, in order to make it generally effective, it was mixed with hydrogen peroxide. The combination of H_2O_2 with HCl was especially effective in clearing the steam-water washes for filtration.

Ammonium carbonate was a fairly effective reagent. It depends on the oxygen of the air for its oxidizing action, corroding copper and putting it into solution as a blue complex, and oxidizing uranium to the six-valent state and putting it into solution as a complex carbonate. The latter action was not as effective as had been intended.

In the experiments in the Alpha plant it was shown that hot air at 300°C to 400°C oxidized the black scale on copper sheets of the liner and collector very effectively so that it could then be readily removed by solvents. This technique was not used in Berkeley.

IV. EQUIPMENT USED TO PERFORM THE RECOVERY STUDIES

14. Equipment Used to Perform the Recovery Studies.

The basic piece of equipment was a large framework (See Figure 2) and collecting basin. The framework was of welded construction, approximately 20 feet high, with a monel metal catch basin, 6 feet by 10 feet by 6" deep, placed high enough to permit standard 50-gallon crocks on dollies to be rolled underneath the basin. The frame was so constructed as to have a clear vertical height of about 15 feet in which the liner could be slung. The width of the unobstructed space

21

RECOVERY WASH-RACK

Figure 2

22

was about 42", and at the four corners of the space vertical angle irons served as supports for demountable walls and ends. These walls were made of panels of thin Masonite, properly overlapped for drainage and painted white with "Amercoat 33" paint. Corner guards were mounted where necessary to prevent losses. During testing of the enclosure with water it was found necessary to provide splash panels which rested at the edge of the basin and leaned against the panels of the frame. For operations on a source the front panel was a single piece with a box-like extension to contain protruding parts of the source. The frame was equipped with two runways, one 6 feet, 6 inches above the other to provide access to the upper sides and ends. These were welded ladders running up on each side. Handrails and guards were provided. Two sets of plumbing were routed to each side of the framework from the rear, and these were fitted with valves, rubber hoses, and plastic nozzles. Floodlights were attached to the top of the frame. After several runs had been complete a motor-driven cable hoist was installed on the top crosspiece to make work on the sources and receiver less difficult.

The entire framework was painted with gray glyptal, a coating which was found to be easily washed and quite resistant.

The monel metal basin had a gentle slope toward center and was provided there with a drain. The drain led to a loose three inch diameter pipe which delivered solutions to the side of the basin. This was used for all the work with recirculating solutions.

The equipment and supplies in use during the recovery operations are listed in Table 2.

23
TABLE 2EQUIPMENT AND SUPPLIES FOR
RECOVERY OPERATIONS

Quantity	Item	Drawing Number
1	"A" Frame	2E 5135A
1	Monel Basin, 6 ft. x 10 ft. x 6" with drain pipe	2E 5154 2F 4522
20	Masonite side panels, 1/4" thick	2E 5114, 5124
1	Front panel, for use with source	2E 5583
1	Hoist, motor, 1.5 tons capacity	
3	Crocks, porcelain 50 gal.	
2	Crocks, porcelain 40 gal.	
3	Crocks, porcelain, 5 and 10 gal.	
3	Tanks, stainless steel, 50 gallon	2E 5754
1	Tank, stainless steel for recirculating line	
1	Tank, vacuum, 20 gallon	2E 5664, 5673, 5683
6	Dollies, steel, with casters	2E 5864
5	Inserts, with hooks, to lift crocks	2E 5553
1	Spreader Ring to guide cables around crocks	2E 5233, 5222, 5251
2	Vats, stainless steel, rectangular 12" x 16" x 24" deep, for soaking parts	
3	Pans, stainless steel	2E 5212
2	Pans, stainless steel, perforated	2E 5202
2	Screen bottom stainless steel beakers for nuts and small items	2E 5621
1	Nash All-Glass Pump, 1 HP, 3400 r.p.m. direct drive	
2	Screenholders and screens to keep solids out of pump	
1	Pitcher, stainless steel, 10 liter	2B 5254
1	Dipper, stainless steel, 2 liter	2E 5543
2	Funnels, Buchner, 9.5 inch	
1	Scales, 500 lb. Fairbanks. Sensitivity 1/4 lb.	
1	Scales, 20 kilo. Sensitivity 1 gm.	
1	Stirring Paddle, stainless steel	2E 5173
1 set	Handling Equipment for the Source	2E 5861, 5894-1, 5894-2 5904
1	Differential Chain Block	
25	Brushes, all kinds, wire and bristle, varnished handles	
10 pr.	Rubber gloves, size 9.5 and 10	
1 roll	Waxed Kraft Paper, 36" width	
1	Beta Particle Counter and Tube, Cyclotron Specialties Company	
	Cables, several lengths and types	
	Chemical glassware, trays, spoons, spatulas, glass filter cloth, filter paper, thermometers, mirror	
	Chemicals: Ammonium carbonate, hydrochloric acid, hydrogen peroxide, oxalic acid, citric acid	
	Copper tubing heating coil for hot water	
1	Kerrick Kleaner, steam generator, Model JGES	
	Portable Floodlights	
1	Steam Plant, available for the last half of the work.	

24

V.

RECOVERY OPERATIONS

15. Operations as a Whole (See also section 4)

A complete cycle of recovery operations required thirty-six hours. Work with the Calutron was usually held up until the collector had been cleaned and the liner could be removed, a total time of about 10 hours. Beta counting of the tank required six hours, chemical cleaning the same, cleaning of a source 8 to 12 hours, a liner 11 hours, and a collector 7 hours.

The chemical crew which was directly concerned with the operations was composed of Oscar Cook, Will D. Phillips, Peter Yankovich, Randall Ross and the writer. Design was largely done by Ralph Peters. Assay work was conducted by Rayburn Dunn and chemical analysis by Thomas Pillsbury. Charge material was supplied by Roy Krohn and Andrew Stein. Much assistance was obtained from operating, maintenance, and repair personnel.

16. Preliminary Operations.

The sequence of preliminary and transfer operations was (1) laying down waxed paper to collect material dropped from the source, removal of the source from the Calutron tank, attachment of supports and a sheet metal pan underneath, closing of the port in the door with a blank plate, and transfer to the wash rack, (2) removal of the collector and transfer to a special dolly, closing the port in the door with a blank plate, gathering up of loose material before further transfer, transfer to the collector service area, removal of the carbon pieces and collector cans, wrapping and identifying each separately, wrapping the whole collector frame with paper, and setting it aside, (3) loosening the door, removal of the blank plates, attachment of special hooks to

25

carry the door and liner, removal from the Calutron tank as soon as the wash rack was cleared of the source, transfer to the wash rack, the path of travel being covered with wax paper, and (4) operations on the Calutron tank. At least one member of the chemical recovery group was on hand for each operation. The transfers were accomplished with a large overhead hoist.

A search was made during and immediately after transfer operations for material which had been dropped, or lost to tools, gloves, and other objects.

17. Recovery Operations on a Source.

After the source was transferred to the wash rack, the side shields were removed, and the charge bottles containing the UO_2 were taken out and weighed. The Masonite panels were then built up, and the steam wash with either the Kerrick Kleaner or the steam plant facilities was begun. Steaming was continued for from 8 to 13 minutes, and 15 to 20 gallons of wash were collected. The source was then disassembled to a point where solder joints would next have to be taken apart, and all the parts were laid down on the monel basin. Steaming and washing were then continued for a short while. The container of solution was then moved from its position under the basin, and a sloping drain pipe was attached. This drain pipe led to a 30-gallon stainless steel tank. A fine sieve was placed at the mouth of the pipe to separate solids from the liquid delivered to the tank. This served to protect the pump. The tank was connected to a Nash All-Glass pump, and this was in turn connected to one of the piping systems for delivery to nozzles on each side. Five percent ammonium carbonate solutions was then made up and delivered to

26

the nozzles at a pressure of 15 lbs./sq.in. The whole source in its stripped condition was then sprayed and scrubbed. The solution was adequately aerated by spraying and draining. Many different bristle and wire brushes were used for the scrubbing. The solution was strengthened with more solid ammonium carbonate as it became weak through volatilization of both components. The Masonite walls were dismantled as the work progressed downward. The scrubbing of the parts in the basin then completed work on the source.

Parts were given a hot water rinse and allowed to drain. All parts of the source which were not put back into service were set aside for inspection and salvage.

For work with the ammonium carbonate solution small ammonia masks were available to the operators. It was also necessary to use plastic face shields, rubber aprons, sleeve gauntlets, and white cloth coats.

18. Recovery Operations on a Liner.

After suitable mounting of the wash rack side walls and splash panels, the liner was thoroughly rinsed with the Kerrick Kleaner, or with steam from the steam plant and water from the spraying system for approximately 15 minutes. Then the walls were taken down, and the liner was dismantled. The detached parts were laid on the monel basin. Rinsing and steaming were then continued for a few minutes, and the collected solution was taken away.

Cleaning scale from the framework and side-panels of the liner was done with brushes, the 5% ammonium carbonate solution, and with a special abrasive paper which would not disintegrate under water. A rotary wire brush cleaner was not found to be satisfactory. The parts and carbon pieces on the basin were finally scrubbed, then rinsed, and the solution

27

was placed in one container. The operations on the liner proved to be difficult and time-consuming.

19. Recovery Operations on a Collector.

The collector framework was moved from the collector service area to the wash rack when the liner was removed from the rack. There was very little to take apart on the collector after the removal of the carbon pieces and cans had been done in the service area. The collector also was not cleaned with steam and water since such an operation would have been fruitless. Two methods were applied. A dry cleaning, with a light hammer, light anvil, and chippers was effective in removing scales thicker than 0.005", but was ineffective on other scales. The most successful method used in Berkeley was continuous spraying of the collector metal parts with 5% ammonium carbonate for about four hours, with occasional light brushing to remove the scale as it was loosened. Finally wire brushes were applied in earnest, in conjunction with continued spraying of the ammonium carbonate solution. Results of the cleaning operations were only fair. A nitric acid treatment was not used by us, but was successfully applied to experimental cleanups at Y-12.

Reassembly of a collector was done with new carbon pieces.

20. Recovery Operations on a Calutron Tank.

The tank walls of XAH-2 were sandblasted prior to Run 33. At the end of this run the walls were given a beta-particle count at 1-foot intervals. This was repeated at the end of Run 34. The results showed that a fair amount of material had escaped to the tank walls, and so an attempt was made to wash the walls with hot water and cloths, but this was not effective. The walls were too rough.

28

It appeared from work on the faceplate during a thorough cleanup of a source that it was necessary to remove rust from the iron if it was hoped to remove the uranium. A study of rust removal by various organic acids and by HCl was therefore made on rusty test samples of mild steel. The results of the study were that 0.5 molar (or about 10%) citric acid and a 0.5 molar HCl solution were equally effective in rust removal. The HCl solution is satisfactory only if the corrosion of iron itself is inhibited with organic amines, such as aniline. The inhibited hydrochloric acid solutions were commercially available. Buffered citric acid solutions were less effective than plain citric acid. Citric acid was chosen for the work at Berkeley, while subsequently the work at Y-12 Alpha-II plant was done with the inhibited hydrochloric acid solutions.

For the second recovery study the cycle of operations on the tank consisted of a sandblasting prior to Run 36, then nothing except pickup of loose material until the end of Run 45. At the end of this run, the tank was washed for 1 hour with the walls at 50°C, with 10% citric acid solution, then rinsed and dried.

21. Salvage Operations.

In the second recovery study 57 groups of items were collected. Twenty groups of carbon parts and eight miscellaneous combustible items were ashed and analyzed. Twenty-one metal items were cleaned in either ammonium carbonate or HCl-H₂O₂ solutions. The parts were scanned with the beta-particle counter after cleaning, and other unsalvaged items were similarly scanned. Brushes were scanned with the beta counter, then cleaned and scanned a second time. The amount left was estimated at 2 grams. Gloves were washed and found to be sufficiently clean. The wash

29

rack required no salvage operations.

22. Treatment of Solutions and Sludges.

The steam-water washes were collected without separation of solids in a single crock and were treated with about 1 gallon of concentrated hydrochloric acid and several pounds of 30% hydrogen peroxide, and heated to about 80°C by means of a copper coil through which hot water was run. This resulted in a mixture composed of a bright yellow-green solution and carbonaceous solids which readily settled out. The mixture was filtered through glass cloth. The solution was accurately weighed and then sampled. The solid was taken for analysis as a whole.

The ammonium carbonate mixtures were filtered without further chemical treatment. The clear filtered solution was weighed accurately and then sampled.

Weighing. All weighings of filtrates was done on a 500-lb. scale which had been calibrated by the California State Bureau of Weights and Measures at the start of the recovery work and which was sensitive to 2 oz. Weighings were reproducible to within 4 oz. by different operators in tests in which the crock and its support were lifted off the balance and replaced by a second and then a third operator.

Sampling. The filtrates were thoroughly stirred before being sampled. All samples were in 2.5 liter bottles, and were properly identified. All solids were treated as a whole.

Volumes and Weights of Solutions and Solids.

The treated filtrates from the steam-water washes for the source and liner averaged 40 and 38 gallons, respectively. The filtered ammonium carbonate solutions from the source, liner, and receiver averaged 51, 70

30

and 44 gallons, respectively. The citric acid tank wash solution was 33 gallons in volume, after filtration. The dry weight of the solids from the various filtrations ranged between 35 grams and 150 grams. These solids ranged in carbon content from 24 to 54 grams. The total amount of carbon found was in all cases much less than would have been guessed from the appearance of the various deposits. The reason was that most of the uranium deposits, even of fairly high chloride content, had a black appearance.

VI: DETERMINATION OF QUANTITIES. UNCERTAINTIES AND ERRORS.

23. Normal Uranium Input.

Since the amount of uranium which entered the recovery work was determined by weight loss of the charge bottles, corrected for volatilization of material other than UCl_4 , careful attention was placed on the iron, chromium, nickel, and HCl content of the UCl_4 . Careful consideration was likewise given to the weight changes of the stainless steel bottles containing the charge and to the consequence of these weight changes on the determination of UCl_4 . The reason for the choice of this method of determination of uranium input was given in Section 3, and important results on the Fe-Mi-Cr-HCl balance were stated in Section 9.

The total amount of UCl_4 which was handled in Recovery Studies I and II was 150 Kg, and the total weight loss was approximately 32 Kg. Since the weight loss was only one-fifth of the total weight of UCl_4 , it is clear that errors in the analyses before and after would be magnified by a factor of 5. The truth of this was shown by an attempted chemical balance which covered 79 Kg. out of the 111 Kg. of UCl_4 which went into

31

Study II. Sixty-four Kg. were left over, representing $5/6$ of the whole amount. The uranium determined by difference of the analytical results before and after was 300 grams more than that which the weight loss method allowed. This was an error of 2%, but would be accounted for by errors as small as 0.18% in the analytical results. The chloride difference was only 70 grams more than the weight loss allowed, or 0.8%. When divided by the factor 12, for twice the ratio of the amount put in to that used, the minimum error is less than the ordinary analytical error in the determination of Cl.

The weight loss from a charge bottle consists of the weight of the HCl, FeCl_3 , and small amounts of NiCl_2 given off as well as the very large amount of UCl_4 . No correction need be made for the UCl_3 and UCl_5 content, since the charge was free of these components. The correction to the gross weight loss on account of the volatilization of these substances was 2.2% (See Ref. 4). The uncertainty in this correction was estimated as less than 0.5%.

It was concluded that the uranium input in Study I was 5566 gm. $\pm$ 50 gm., the uncertainty therefore being $\pm$ 0.9%. For Study II the uranium input was determined as 14568 gm. $\pm$ 100 gm., and the uncertainty was $\pm$ 0.7%. These uncertainties were the best which could be decided on after very careful consideration of the problems which were involved, and represent the best we could have done under the circumstances. The only way of lowering the uncertainty would be to use a larger fraction of the UCl_4 . With utilization of two-thirds of the UCl_4 , the uncertainties would have amounted to 0.2%. It is, however, difficult to reduce the uncertainty to

32

less than 0.15% on account of unavoidable errors in the chemical analyses.

Additional light has been thrown on the determination of the UO_2 loss from charge bottles by weight loss. A very careful set of material balances on the charge material before and after a run were made in March, 1945 (Ref. 13) in which the weight loss in both nickel and stainless steel charge bottles was compared with the results of chemical analysis. The charge contained in the bottles ranged from 120 to 300 grams, and the fraction sublimed ranged from 12% to 80%. The uranium sublimed, as determined by chemical analysis, was set equal to 1.000. On this basis, the weight loss determinations of uranium sublimed from nickel containers were 1.002, 1.006, 1.002, 1.002, and 1.000, and from the stainless steel containers 1.02, 1.014, and 1.027. These results support the conclusions made above, and the size of the correction which was applied.

24. Normal Uranium Found in Solutions and Solids.

The determination of uranium in the various solutions and solids was made by analytical methods which had shown their usefulness and accuracy.

The steam-water wash solutions were analyzed both volumetrically and gravimetrically. The volumetric method consisted of reduction of the solution by means of a silver reductor, after the excess H_2O_2 was removed, followed by titration with ceric sulfate and ferrous sulfate. The gravimetric method consisted of the separation of iron by means of ammonium carbonate and the subsequent precipitation of uranium by means of carbonate free ammonium hydroxide. The two methods together could be counted on to determine the uranium to within 0.1%.

33

The ammonium carbonate solutions were analyzed by ether extraction of the uranium and determination of the uranium gravimetrically with a double cupferron procedure.

Solids were dried and then ashed in entirety. The ash was then ground and sampled. The sample was treated with a mixture of HCl-HNO_3 , and the residue which remained was fused with NaHSO_4 . The solutions resulting from the two treatments were then combined and analyzed gravimetrically. The residue from the NaHSO_4 treatment usually showed a high beta-particle activity, but this was ascribed without proof to ThO_2 or other material, and not to uranium left in the residues.

Carbon pieces which were submitted for assay were entirely ashed, and then analyzed by a modified uranium oxalate method. The superior ether extraction-double cupferron method was not in use by the assay group at the time.

Weighings of the recovery solutions were made to within 1 part in 1000.

The solutions submitted for analysis contained primarily Fe, Cu, and Ni along with the Uranium. The steam-water wash solutions averaged 3.9, 0.3, 1.5 and 0.05 gm./liter for U, Fe, Cu, and Ni, respectively, from the sources, and 9.0, 0.4, 1.8, and 0.03 gm./liter from the liners. The ammonium carbonate solutions were very rich in copper, but were free of iron. The uranium and copper content, respectively for the solutions from the sources were 0.26 and 6.2 gm./liter, from the liners 0.32 and 6.2 gm./liter, and from the collectors 0.11 and 5.0 gm./liter. The solution from the citric acid tank wash contained 98 gm. of U, 1542 gm. of Fe, 50 gm. of Cu, and 298 gm. of Cl in 33 gallons.

The uranium content of the solids was ordinarily not an important part

34

of the total uranium. The amounts of uranium found in the solids ranged from 4.0 to 470 gm., the latter being 11% of the total uranium for the particular run.

It was found to be very necessary to consider each analytical result obtained in the light of the origin of the sample and the consistency of the result, both internally and by comparison with analyses on similar materials. Two very important errors were found by this means and reanalysis was required to correct the results. The most probable overall uncertainty of the analytical results was between 0.1 and 0.2%.

25. Normal Uranium in the Salvage.

The salvage pile was so handled as to keep it an unimportant part of the work in terms of uranium content. The normal uranium recovered from the salvaged items in Study I was 0.2% of the whole, and in Study II 0.59% of the whole. The analytical methods applied were sufficiently accurate, being 1% in error in quantities measured. Loss in items not considered salvageable was estimated to be not over 20 grams, or 0.1% of the total uranium. There may have been larger unknown losses.

26. Enhanced Uranium. The Product.

The measurement of the enhanced uranium by the assay group usually involved errors as small as 1% to 5% for the determination of the total uranium involved. There is no need to consider here the degree of enhancement of this material. The uranium involved in this product amounted to 0.43% of the whole amount.

27. Impoverished Uranium.

Insofar as the enhanced uranium is a product of the operations, so also is the impoverished uranium. The total amount of both which had to

35

be reclaimed from the carbon pieces in the collector and from the cans was 1491 gm. in the two recovery studies. This is about 7.5% of the total. The overall error in the determination of the uranium involved in this material was about 2%. Approximately 500 grams of material which should have been found on these parts, as determined by integration of the meter readings in the Calutron control room, was scattered to other parts of the receiver. Slightly more than this amount of material was found on the collector, namely 515 grams altogether. The average U_{235} content of this material was 0.53% instead of the normal 0.64%.

28. Uranium Located but not Gathered.

The uranium which was not gathered was in some cases located by beta-particle counts. Alpha particle counting, which would be accurate for extremely thin films was excluded both on account of the thickness of the films involved and on account of the short range of the alpha particles in air. The methods used in Beta-counting are explained in Section 35. (See also Ref. 6, 7, and 8)

Our chief concern in applying the beta count to apparatus was to apply it to apparatus which was either new at the start of the work or was sand-blasted clean. Therefore both the liner and the tank were counted at the end of Runs 33 and 34 of Series I. The new source, liner, and collector were counted also at the end of Series II.

The total amount which it was estimated was located but not gathered on the major units during Recovery Study II was 194 grams, or about 1.3% of the total uranium. When this number was corrected for the cross-check which was available between beta-count and chemical analysis, the estimate was increased to 300 grams, with an uncertainty of 100 gm. or 0.7% of the total uranium used. The numerical results, without statement of the un-

36

TABLE 3

DETERMINATION OF THE APPROXIMATE
INCREASE IN THE URANIUM CONTENT
OF UNITS BY BETA COUNT
(all numbers = grams U)

	Run No.	Source		Liner		Collector		Tank XAH-2	
		Gregor	Goofy	UCRL	Westing- house	Dorothy	Alpha- gloria		
Recovery Study I	Prior Value			0		0		32	
	After 33	?		29		?		64	
	After 34	?		44		?		100	
	Increase	?		44		?		68	Total 112
Recovery Study II	Prior Value	?	0		0	?	0	15	
	After 36	?	1		69	?	100	42	
	After 45	?	62		80	75	30	37	
	Increase	?	62		80	?	30	22	Total 194
	Overall Change	?	62	44	80	75	30	90	Total 379

37

certainty of each are summarized in Table 3.

29. Uranium Lost.

It is not possible to make any estimates about the amount of uranium which was lost, except by the difference between the final recovery value and 100%. This will be taken up in Section 32. All efforts were bent toward reducing known losses, excepting those measured by beta-counting, to zero. Material which is lost, insofar as it is not in a recovery container but still on a unit, is of less importance if the units see repeated use and then have a thorough cleaning before going to the discard pile.

30. Variation of the Uranium Content of the Calutron Equipment.

For an experimental study of recovery one of the real sources of concern is the variation in the equipment itself. For those items which were new or sandblasted it was possible to make a fair, but low, estimate of the amount held up (see sections 28 and 35). The difficulty of cleaning a source, the porosity of its castings, and the relative ease of diffusion of uranium into copper at 400°C or above (Ref. 14) all make it impossible to form a good estimate of the material held up by this unit. It may be estimated that a well cleaned source will vary in its uranium content by 50 grams from run to run. It is known that a well cleaned source may still contain at least 144 grams of uranium (see section 34).

The best we could do was to try to do the cleaning the same way each time. The extent to which this was possible, as checked on "old" equipment in subsequent work in the Alpha-II plant at Y-12, is stated in Section 35. The conditions were better there for doing the cleaning the same way each time, however.

38

For Recovery Studies I and II together, which included the removal of 20 Kg. of uranium from two sources, two collectors, two liners, and one tank, the uncertainty in identity of the equipment before and after the studies amounts to an estimated loss of 100 grams with a variation of 50 grams. This estimate is over and above the amount actually accounted for by beta-counts.

31. Recovery Results.

The results of the recovery operations have been summarized in Table 4. The maximum recovery claimed is 95%. The breakdown of the recovery results is given in Table 5, to show the input for each run, the size of the correction to the charge bottle weight loss, and the distribution and effectiveness of removal of uranium from the various units.

Accountability. The overall loss figures for Studies I and II together is 1110 grams, or 5.5%. The uncertainties mentioned in sections 23 through 30 may be summarized as $\pm 0.8\%$ in the uranium input, $\pm 0.2\%$ in the analytical work on the solutions and solids, $\pm 0.1\%$ in the work on salvaged items, $\pm 0.1\%$ in weighings in the recovery department, $\pm 0.2\%$ in the assay department and $\pm 0.25\%$ in the holdup on units. In addition, the holdup on units was estimated to increase by a total of 100 grams, or 0.5%. The beta count figures, corrected as explained previously (and in more detail in Ref. 4) account for $2.5\% \pm 0.8\%$. The overall uncertainty, defined as the square root of the sum of the squares of the individual uncertainties, is therefore $\pm 1.2\%$.

Since the loss figures of 5.5% is the sum of the values for the material not accounted for plus that accounted for by beta counts, plus

39

the increase in holdup, the percentage not accounted for is $2.5\% \pm 1.2\%$.

It may be concluded therefore that $97.5\% \pm 1.2\%$ of the uranium was accounted for.

Consistency of Performance. It may be noticed in Table 4 that there is a remarkable consistency in the percentage recovery of the uranium from the major units. The liners were all excellent in preventing loss of uranium to the tank walls, and therefore they fulfilled their purpose in this respect very well. The amount escaping the liner to the tank walls ranged from $2\% \pm 1\%$ in Study I through 1.2% with the Westinghouse liner in Study II, to 0.27% for the slightly different Alpha I/5 liner.

32. Consequences of the Final Results.

The consequences of the recovery studies were (1) the conclusion that the losses from the liners to the tank walls were very low, probably as low as 0.5% provided there would be no mechanical loss to the tank during removal, (2) that the overall loss reported for the work in Berkeley was considered low enough to regard the Alpha-II plant at Y-12 as being capable of being used for charge material with enhanced U_{235} content, and (3) that the utilization efficiency of the U_{235} was between 50 and 60%. The actual utilization efficiency figure is 56% when based on an assumed storage of 1500 grams of material having a U_{235} content of 0.05% or less, the storage of 515 grams of material taken off collector surfaces and having an average U_{235} content of 0.53% , an assumed loss of 500 grams of normal material having a U_{235} content of 0.64% , and a production of 85 grams of material containing about $10\% U_{235}$. (See Section 33 for utilization efficiency studies at the Y-12 Site and for definition of the term.)

40

TABLE 4

SUMMARY OF THE RECOVERY STUDIES
(Chemical Recovery)

Recovery Study			
	I	II	III
XAH-2 Runs	33-4	36-45	54-57
U Input	5566 gm	14568 gm	4350 gm
U Recovered	5179 gm	13845 gm	X
% Recovered	93.0	95.0	X
Recovery Breakdown by Units % of Input			
Source	37.5%	37.2%	X
Liner	45.2%	46.1%	44.7%
Collector	10.0%	10.0%	X
Tank	X	1.2%	0.27%
Salvage	0.3%	0.6%	1.3%

TABLE 5

URANIUM RECOVERY, CLASSIFIED BY OPERATIONS
All quantities given as grams

XAH-2 Run Number

Unit	Description	33	34	36	37	38	39	40	41	42-3	44	45	Final Cleanup
	<u>Input</u>												
	Charge Bottle Weight Loss	4294	4790	5794	2066	1819	1241	1209	3098	2170	1418	4963	
	Corrected Weight Loss	4206	4677	5720	2195	1845	1195	1155	3050	1960	1380	4933	
	Uranium Input, Calculated	2636	2930	3590	1250	1090	750	720	1910	1225	870	3090	
<u>Source</u>	<u>Recovery</u>												
	Steam-Water Wash Solution	663	905	1147.1	←		1417.→		←988.3→		81.0	1084.→	-
	" " " Solids	4.5	13	4.05	←		11.5→		-	-	12.7	71.4	-
	Ammonium Carbonate Solution	231	171.8	52.9	39.2	-	-	-	-	-	30.81	80.0	-
	" " " Solids	24.2	57.3	14.7	25.5	-	-	-	-	-	53.64	56.97	-
	Source Carbons	1.1	22.9	-	←		20.8→		-	-	-	34.5	53.2
	Source Pre-Cleaning*	-	-	-	-	-	-	-	-	-	-	-	143.1
<u>Liner</u>	Steam-Water Wash Solution	1094.	1183.	1576.	←		1077.2→		←1477.8→			1328→	-
	" " " Solids	2.4	9.1	71.6	←		214.4→		-	-	-	470.7→	-
	Ammonium Carbonate Solution	54.3	80.3	75.2	-	-	-	-	-	-	-	-	137.6
	" " " Solids	26.9	30.5	39.6	-	-	-	-	-	-	-	-	176.6
	Liner Carbons	1.6	31.1	-	-	-	-	-	-	-	-	-	69
<u>Collector</u>	Carbons Impoverished U.**	200.	222.	335.9	←		247.1→		151.9		249.0	→	-
	Cans and Carbons, Enhanced U.	9.0	9.6	22.7	←		16.6→		8.5	1.32	18.3	→	-
	Receiver Carbon Salvage**	-	-	-	-	-	-	-	-	-	-	-	31.6
	Receiver Dry Cleaning**	-	-	-	←		76.3→		-	-	-	-	-
	Ammonium Carbonate Solution**	25.0	14.8	14.2	-	-	-	-	←40.6→				14.9
	" " " Solids**	43.7	30.0	26.9	-	-	-	-	←193.7→				13.5
<u>Tank</u>	Tank Sweeping	0.8	5.2	3.9	←		6.6→		-	-	-	-	-
	Citric Acid Cleaning Solution	-	-	-	-	-	-	-	-	-	-	-	164.0
<u>Salvage</u>	Salvage	11.9	0.2	10.1	←		1.5→		←12.6→				62.0

*See Section 34.

**These materials are subject to
storage due to U₂₃₅ impoverishment.

43

Appendix I.

33. Study of Recovery in Alpha-II at Oak Ridge, Tenn.--Y-12.

Recovery studies were conducted in the Alpha-II plant in the Y-12 area simultaneously with the work stated in this paper. These studies were conducted under less favorable circumstances than the studies in Berkeley, and the results were less satisfactory. Therefore in March, 1945, a decision was reached to do a thoroughgoing job of recovery on an experimental basis with the selected units in Track 6. The sources, liners, and collectors were identical to the production units except for modifications desired for the experiments. The work was undertaken in May and June, 1945, and the writer went to Oak Ridge to be of assistance in the studies. The studies had as their goal the measurement of utilization efficiency, a somewhat broader goal than measurement of recovery alone, since it includes determination of the U_{235} content of all samples. (See Ref. 5).

Utilization efficiency may be defined as the ratio of the amount of the U_{235} which finds its way into the product divided by the sum of the amount in the product plus the U_{235} in the material which cannot be accounted for and is considered lost plus the U_{235} in the material which is impoverished to a point where it is most economical to withdraw the material from the cycle of operations. For the work in Berkeley the best figures which can be given for this quantity is 56%. For the recovery studies in the Alpha-II plant, Track 6, the results reported for this quantity were quite interesting but would require more space to tell here than should be taken.

The studies consisted of eight runs, four without recovery liners and four with. The plan was to clean each of the four tanks, then perform a non-liner run in it, clean the tank again, and perform a liner run in it,

44

finally cleaning the tank a third time. The cleaning of the tanks was done with inhibited HCl solutions. The results with the non-liner runs all gave recoveries above 100%, and are thus not valid for the estimation of losses. The recovery from the tank walls appears to have been more than was put into the tank during the run. The liner recovery studies were made in the same tanks, and it may be guessed that the tanks would no longer contribute extra amounts of uranium upon being cleaned a third time. The loss determined in the three valid liner runs was 2.2%, 1.7% and 2.2%, with an estimated overall uncertainty of 1%.

It appears that a 2% loss is about the amount which is to be expected in careful recovery studies with Alpha-II equipment. This is in agreement with the conclusion stated in section 31.

Appendix II.

34. Recovery of Uranium from a Source Scheduled for Repair.

At the close of XAH-2 Run 34, the termination of Recovery Study I, the source unit "Gregor" which was used was in need of an insulated bushing replacement in such a spot as required the complete dismantling of the unit. This therefore offered an opportunity for cleaning the unit beyond the point which could be attained in the recovery studies.

"Gregor" had a total of 200 days of service, prior to Recovery Study I. It had then been given three complete cleanings by the methods outlined in Section 7 and 17. These cleanings could not remove all the uranium, however. The castings were observed to be very porous, and the interior parts of the source dirty, scaled, and rusty even after the recovery cleaning process.

To determine the quantity of uranium which could be obtained by cleaning the dismantled parts of the source, the source was disassembled and the parts

45

were scanned with a beta-counter to orient the personnel to the problem, and marked with chalk on those places needing particular attention; then cleaning operations were begun and continued for a week. The parts routinely disassembled from a source during service operations were not included in the study. Altogether 40 parts or sub-assemblies were cleaned. The methods used included (a) scraping, dry and wet, (b) wire-brushing by hand and by air-powered motor, (c) ammonium carbonate spraying, plus brushing, (d) soaking in ammonium carbonate solution for one to two days, then brushing, (e) use of a saturated oxalic acid solution as a rust remover, and (f) use of HCl-H₂O₂ mixtures of various strengths combined with wire-brushing.

It was estimated that between 90% and 95% of the visible surfaces were cleaned to bare metal. Cavities, pits, and cracks were cleaned as well as soaking could clean them. It was found, however, that all castings concerned with the holding of the charge bottles and with conducting the gas to the arcs, and on which the arc products directly played, were still very active in beta-count after being reduced to bare metal. Also having high beta-count activity after cleaning were the areas covered with silver solder. It may be concluded that uranium had diffused into the metals where the temperatures were high. Stainless steel was resistant to such diffusion, however.

The total uranium recovered from the parts of the unit was 143.6 grams. This was distributed as follows:

Exposed areas of the steel faceplate-----	1.2%
Hidden areas of the steel faceplate-----	1.8%
Insulated central framework, consisting of a bronze casting, and its attached heater leads and water lines-----	18.1%
Charge bottle and arc box castings, linings, and heat shields-----	64.4%
Uninsulated bronze support casting, with the attached rectangular casting mounted in front of the arcs, plus the inter-arc fins-----	9.4%
Miscellaneous and salvage-----	4.9%

46

It may be noticed that the largest amount of uranium was associated with the parts the UO_2 had the most contact with at the highest temperature.

The cleaned parts probably still contained from 50 to 100 grams of unreclaimed uranium.

35. The Beta-Count.

Radiation counting instruments were used in the UCRL Alpha-II Recovery Studies (Section 28, Ref. 1,2,3,4,6) in such a manner as to determine the amount of uranium left on surfaces with a probable error of about 30%. Subsequent work in the Alpha-II plant in the Y-12 area showed that the determination of uranium by the method gave estimated probable errors of 50%, for the usual work, or 25% with units primarily composed of flat or smoothly curved surface. It was found that the results of the counting surveys generally appeared to have less error than this, but the chemical check obtained after careful beta-counts at the end of Run 45 (Recovery Study II-UCRL) dictated considerable caution in accepting beta-count values without revising them upwards. The difference between two beta-counts, before and after citric acid cleaning of the tank, was 92 grams of uranium, while the chemical analysis showed 163 grams.

The basis of the beta-particle estimation of uranium is the following: U_{235} loses an alpha particle, becoming UX_1 , an isotope of Thorium. This emits a moderately energetic electron and becomes UX_2 , which then emits a very energetic electron and becomes UZ_{234} . This very energetic electron is the one on which the beta-count work depends. The other chief isotope of uranium, U_{238} , loses an alpha particle, becoming UY_{234} . This emits a moderately energetic electron and becomes Pa_{234} , a fairly stable product. The very energetic electron from UX_2 is separated from the two other less

energetic electrons by means of a very thin film of material, such as 10 mils of mica or 2 mils of copper. For the energetic electron the absorption in matter is not so rapid. A 10 mil sheet of copper cuts down the activity by a factor of 4.2. (See also Ref. 8)

For chemical recovery in alpha units, using the beta-count on U_{238} is very logical. It is the chief material one is looking for. The energetic character of the electron from U_{238} is also required to give any valid estimates from materials of varying thickness, some of which have to be viewed with the counter at a distance of 6" to 12". The chief disadvantage is the fact that pure U_{238} has no beta activity, but slowly builds it up when separated from its "daughters". It one assumes complete separation, the rate of buildup (Ref. 6) is about 8% per day at first, tapering to 4% by the 15th day, and 1% by the 25th day, thereafter being very small. The mathematical curve for the rate of buildup was verified by taking samples from various parts of the equipment after runs were completed. It was also shown that in the sublimation of UO_2 at 450° to 500°C, the beta-particle activity of the sublimate was reduced to a very small fraction of the equilibrium activity. This leads to the conclusion that it is desirable to apply the beta-count to pieces of equipment only after waiting a period of days. Sometimes this proved to be practical, other times not.

The limitations on the use of beta-counts for determining the amount of uranium lay in: (1) the variable thickness of scales; (2) the penetration of uranium into hot metals; (3) inability to place the viewing face of the counter tube directly over the surface; (4) uncertainty concerning the "age" of the U_{238} ; provided that the "age" was less than 15 days; and (5) the decrease in the energetic beta activity per gram of total uranium as the

48

U_{235} content increased. With respect to these limitations it was found (1) that a 10 mil thickness of scale on receiver surfaces required multiplying the beta-count determination by a factor of 2, (2) nothing could be done about penetration, (3) that a one-spot view, correcting the reading as one would for a light source, had to suffice, (4) that the "age" determination had to be ignored more often than used, since many of the units employed were in use for periods of time up to 200 days before the recovery work was started, and (5) that the variation of the count with U_{235} content could be completely accounted for. All these limitations led to the statements regarding uncertainty of the method given in the first paragraph above.

In the UCNL work it was possible to use ordinary counter circuits, with an integration of the count being made over fairly satisfactory time intervals. Such counters were not usable in the work at Y-12, however, since the distance from the wire to wall for the counter tubes was too large, and the smaller counter tubes which would work were attachable only to a rate-meter circuit. Integration counters were also not usable since the "background" in the region of the track was double that outside the track room and extremely variable with sparking. It was necessary to use a rate-meter circuit throughout, to hold the counter tube in the magnetic field with the wire at right angles to the lines of force of the magnetic field, and to wait for time intervals in which the spark activity was small. It was also necessary to check the rate-meter reading with a standard sample for every reading made in the magnetic field.

Estimates based on beta-counts were identical when made either with a counter held still over representative samples areas of a surface, or moved over the area continuously in a manner such as would integrate the whole

49

surface. In all the work we gradually swung over completely to observations made with moving counter tubes. These were frequently more rapid.

Results on beta-count surveys of the recovery equipment have already been summarized for the UCHL work (Section 28). In the work at X-12, the beta-count surveys (Ref. 7) were made with the purpose of demonstrating the fact that the equipment was left in approximately the same condition with respect to uranium content after the work as it was before a recovery run. The results were as follows:

GRAMS OF URANIUM DETERMINED BY BETA-COUNT				
	Non-Liner Recovery Study (JM-4)		Liner Recovery Study (JM-8)	
	BEFORE	AFTER	BEFORE	AFTER
	Tank 687		Tank 687	
Collector end				
and top	188	167	167	159
Balance	7	11	11	7
Manifolds	71	22	22	21
	Source 536		Source 181	
Removable				
Parts	5	5	5	4
Fixed Part	154	99	61	61
	Liner 56		Liner 244	
Total	15	11	18	19
	Collector 197		Collector 7	
Total	2	2	9	9
Totals	442 gm.	317 gm.	293 gm.	286 gm.

The beta-count results for the Liner Recovery Study (JM-8) argue well for the statement that the equipment was left in the same condition as it was found, within the ability to tell both from visual observation and from the beta-count.

50

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1-50

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53117