~~SECRET~~

Y-697

**THE SEPARATION AND
COLLECTION OF U-236
BY THE
ELECTROMAGNETIC
PROCESS**

BY

**H. W. Savage
P. E. Wilkinson**

**OAK RIDGE NATIONAL LABORATORY
Y-12 AREA
OPERATED BY
CARBIDE AND CARBON CHEMICALS DIVISION
UNION CARBIDE AND CARBON CORPORATION
OAK RIDGE, TENNESSEE**

~~SECRET~~

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

~~SECRET~~

Index No. Y-697

This document contains 136 pages.

This is 29 of 36, Series A.

Subject Category: Technology-Electromagnetic Process

THE SEPARATION AND COLLECTION OF U-236 BY THE ELECTROMAGNETIC PROCESS

H. W. Savage
P. E. Wilkinson

CLASSIFICATION CANCELLED
DATE - 4 - 6 - 56
For the Atomic Energy Commission

H. F. Canale
Chief, Declassification Branch *RET*

November 30, 1950

ISOTOPE RESEARCH AND PRODUCTION DIVISION
C. P. Keim, Director

OAK RIDGE NATIONAL LABORATORY
Y-12 AREA
CARBIDE AND CARBON CHEMICALS DIVISION
UNION CARBIDE AND CARBON CORPORATION

Oak Ridge, Tennessee

Contract No. W-7405-eng-26

UNCLASSIFIED

RESTRICTED DATA

This document contains restricted data as defined in the Atomic Energy Act of 1946.

~~SECRET~~

CAUTION

This document contains information affecting the National Defense of the United States. Its transmission or the revelation of its contents in any manner to an unauthorized person is prohibited by applicable Federal laws.

~~SECRET~~

Index No. Y-697
Technology Electro-
magnetic Process

Distribution, Series A:

- 1-25. Carbide and Carbon Chemicals Division (Y-12 Area)
- 26-27. Atomic Energy Commission, Washington
- 28. Patent Branch, Washington
- 29-34. Technical Information Service, Oak Ridge
- 35-36. University of California Radiation Laboratory

Carbide and Carbon Chemicals Division (Y-12 Area) internal distribution as follows:

- 1. C. E. Center
- 2. C. E. Larson
- 3. W. B. Humes
- 4. W. D. Lavers
- 5. E. D. Shipley
- 6. A. M. Weinberg
- 7. C. P. Keim
- 8. R. S. Livingston
- 9. G. H. Clewett
- 10. J. M. Herndon
- 11. R. F. Hibbs
- 12. Boyd Weaver
- 13. P. J. Hagelston
- 14. L. E. Burkhart
- 15. C. E. Normand
- 16. C. R. Baldock
- 17. J. R. McNally, Jr.
- 18. H. M. Roth
- 19. J. W. Ruch
- 20. E. G. Struxness
- 21. P. E. Wilkinson
- 22-25. C. R. I. O.

Issuing Office
Plant Records Department, Y-12 Area
Date Issued: 1-9-51

~~SECRET~~

~~SECRET~~

3.

3

ABSTRACT

Six samples of uranium containing about 20% U-236 each have been collected from 11.3 grams of uranium in which the U-236 was 0.82%. A 12-inch radius, high-recovery calutron was designed, tested and used. Details of the experimental, design, calutron operations, chemistry, mass analysis, and health physics programs are included as appendices, and each is summarized in the report.

~~SECRET~~

4

~~SECRET~~

PREFACE

This report describes the work done to collect several samples of uranium enriched in U-236, which began in December, 1947 in the Isotope Research and Production Division.

Because knowledge of the nuclear properties of the U-236 isotope appeared urgent, the problems of methods and equipment for its separation and refinement were subjected to simultaneous design and experimental attack. The concurrent efforts of several groups and many individuals have made it difficult to report the results either chronologically or by subject without appearing to lose some of the significance of the effects of one phase of the program on another. By the device of writing the body of the report as a summary of primary results and experiences and writing the details of each phase in a separate appendix, the authors sincerely hope that most of the successive consequences of ideas have been retained.

Considerable urgency was attached to the project; consequently, the cooperative efforts of the Isotope Production and Development, Isotope Chemistry, Isotope Physics, Engineering, Mass Analysis, and Health Physics Group of Y-12 were required. Their efforts made possible the development and use of a new calutron, chemical techniques applicable to small quantities of uranium, the handling of a hazardous radio-active material, and the successful collection of U-236 in approximately 11 months. To the many people who were engaged in this cooperative effort, every gratitude is extended.

In particular we wish to recognize the contributions of those who were, in general, responsible for one or another of the phases of the project.

~~SECRET~~

~~SECRET~~

H. M. Roth and St. George T. Arnold of the Atomic Energy Commission whose encouragement, advice, liaison work and expediting of both information and the uranium, were invaluable.

The appendices and their authors are listed below:

Appendix A - Methods of Estimation of Product Purity	H. W. Savage P. E. Wilkinson
Appendix B - Preliminary Experimental Program	H. W. Savage P. E. Wilkinson
Appendix C - General Design of the 12-Inch Radius Unit for Uranium	H. W. Savage P. E. Wilkinson
Appendix D - Handling Equipment	H. W. Savage W. G. Cobb
Appendix E - Ion Source Development	H. W. Savage M. J. Skinner G. D. Whitman P. E. Wilkinson
Appendix F- Receiver Development	H. W. Savage M. J. Skinner G. D. Whitman P. E. Wilkinson
Appendix G - Charge Chemistry and Recycle Operations	Boyd Weaver J. L. Williams
Appendix H - Product Chemistry	Boyd Weaver W. C. Davis
Appendix I - Spectrographic Assay	J. R. McNally
Appendix J - Health Physics	E. G. Struxness
Appendix K - Final Runs and Operations	H. W. Savage M. J. Skinner G. D. Whitman P. E. Wilkinson

In addition to those contributing directly to the preparation of this manuscript, numerous others contributed heavily to the successful conclusion of the project.

~~SECRET~~

~~SECRET~~

Our gratitude is extended to Dr. E. D. Shipley for his administrative and technical advice. We want to express our appreciation for the administrative direction of Dr. C. P. Keim; and for his, and Mr. J. E. Keeton's work in obtaining priorities and expediting the work within the Y-12 organization. We want to thank Dr. C. E. Normand for his work on our vacuum problems and other technical and administrative direction.

A large part of the credit for the success of the project belongs to Messrs. Ernest Zurcher, J. S. Culver, W. L. Scott, and G. M. Lindner for their engineering and design work and whole-hearted cooperation in the project.

The product assay work was made possible through the cooperation and efforts of Mr. L. E. Burkhart, Dr. J. R. McNally, Jr., and Mr. D. D. Smith.

The recycle process and product recovery, both essential to the completion of the project, were developed and carried out by Messrs. Boyd Weaver, W. C. Davis, W. A. Burns, J. L. Williams, F. B. Thomas, E. H. Swanson, S. F. Fairbourne, C. F. Harrison, and C. W. Sheridan. The cooperation and effort displayed by this group were major contributions to the successful completion of the project.

The safety of the personnel involved was handled through the efforts of Mr. E. G. Struxness, and Mr. J. H. Hackney.

The authors wish to acknowledge the assistance received from, and the work performed by, Messrs. W. A. Bell, W. W. Davis, L. O. Love, J. W. Raleigh, O. B. Rudolph, K. A. Spainhour, T. A. Starnes, N. A. Stone, and H. E. Walchli both on operation and development of the equipment used on this project. Special recognition is due Messrs. W. G. Cobb, F. R. Duncan, M. J. Skinner,

~~SECRET~~

~~SECRET~~

and G. D. Whitman for their many contributions to the development of the equipment and operational techniques used.

Messrs. J. W. Simmons, N. V. Smith, and Clifford Russell assisted on many of the project details.

Mr. R. F. Hibbs supervised the assay work and contributed to the numerous conferences held.

The appreciation of the authors is extended to Mr. J. R. Patton for extensive revisions and final editing of the manuscript, and R. N. Goslin and Dr. C. E. Normand for their help in this work.

~~SECRET~~

~~SECRET~~

TABLE OF CONTENTS

INTRODUCTION	10
PRELIMINARY CALUTRON SPECIFICATIONS	11
ESTIMATES MADE BY THE Y-12 ISOTOPE RESEARCH AND PRODUCTION DIVISION	13
PRELIMINARY EXPERIMENTAL PROGRAM	14
CHARACTERISTICS OF THE 12-INCH RADIUS UNIT USING UCl_4 CHARGE	15
CHARGE CHEMISTRY	21
PRODUCT RECOVERY	23
HEALTH PHYSICS	25
SPECTROGRAPHIC ASSAY	26
CONCLUSIONS	28
APPENDICES	
A. METHODS OF ESTIMATION OF PRODUCT PURITY	30
Estimate of Collection Rate	31
Effect of Ion Exit Slit Area	31
Effect of Beam Trajectory	31
Effect of 90° Baffle Opening	32
Source Voltage	32
Estimate of Charge Consumption Rate and Process Efficiency	33
Estimates of Product Quantity	34
Estimate of Enhancement	35
B. PRELIMINARY EXPERIMENTAL PROGRAM	38
C. GENERAL DESIGN OF THE 12-INCH RADIUS UNIT FOR URANIUM	47
D. HANDLING EQUIPMENT	56
E. ION SOURCE DEVELOPMENT	58
Support Structure Redesign	59
Vapor Control	59
Oven	61
Arc Casting	64
Charge Bottle	65
Temperature Regulation	67
Electrode Region	68
F. RECEIVER DEVELOPMENT AND BEAM STUDIES	73
Monitoring Receivers	73
Collection Receivers	75
Collection Receiver No. 1	75
Collection Receiver No. 2	77
Receiver Defining Slot Experiments	79
Side Band Shielding	86
Shutters	86
U-236 Pocket	87

~~SECRET~~

~~SECRET~~

APPENDICES (Cont'd)

G.	CHARGE CHEMISTRY AND RECYCLE OPERATIONS.	92
	HCP Process Development.	92
	Radiation Hazard	93
	Process Description.	93
	Calutron Run	94
	Unit Wash.	95
	Uranium Extraction	96
	Salvage.	97
	Run Figures and Recovery Date.	98
H.	CHEMICAL PURIFICATION OF U-236	103
I.	SPECTROGRAPHIC ASSAY OF PRODUCT MATERIAL	107
	Theory of Isotope Shift in Heavy Elements.	107
	Spectro-Isotopic Assay of Samples Enriched in U-236.	107
J.	HEALTH HAZARDS	116
K.	FINAL RUNS AND OPERATIONS.	123
	Collections.	123
	Methods of Estimating Product Purity	129

~~SECRET~~

~~SECRET~~THE SEPARATION AND COLLECTION OF U-236 BY THE ELECTROMAGNETIC PROCESS

INTRODUCTION

Early in December, 1947, the Atomic Energy Commission requested that consideration be given to the collection of one or more samples enriched in U-236 from a small amount of product level uranium which had been bombarded in a Hanford pile. At that time it was estimated that about fourteen grams of uranium would be available and that its isotopic composition would be about one per cent U-236, about seven per cent U-238, and the balance U-235. On the basis of this information, the Y-12 Isotope Research and Production Division was asked to submit a preliminary estimate as to the quantity and quality of product which could be expected, and the quantity of material that could be returned after processing.

At various later dates further information as to the quantity and isotopic composition of the initial charge material became available. Consequently, revised isotopic abundance figures appear at various places in this report which indicate the best information available at the time of that particular phase of the project. The Atomic Energy Commission later specified one milligram of U-236 in a total of 6.67 milligrams of uranium (i.e., 15 per cent), as a minimum product requirement. Also, return to the Atomic Energy Commission of as much as possible of the initial material after processing was requested.

Three primary problems were entailed in making these separations each of which required a detailed solution. These were:

~~SECRET~~

~~SECRET~~

1. Development of calutron equipment capable of handling small amounts of material and from which high recovery could be expected.
2. Development of adequate chemical equipment and techniques for assuring maximum recovery of material and removal of contaminants from the product.
3. Development of techniques for assuring complete protection of personnel from possible radiation hazards.

PRELIMINARY CALUTRON SPECIFICATIONS

The major considerations determining preliminary calutron specifications were: (1) the small amount of U-236 bearing material available, and (2) the desire for maximum recovery.

The belief that the material holdup by the comparatively large surfaces of Beta-type equipment would approximate the total amount available indicated the use of smaller equipment. Also, initial estimates of product quantity and quality based on known operating characteristics of both Alpha and Beta equipment, and on anticipated performance of a still smaller calutron, indicated that several collection runs might be required. These considerations made the problems of recovery and recycle of charge material still more pronounced.

The available amount of 236-bearing uranium made a small calutron further desirable in that only in equipment of reduced size could each

~~SECRET~~

~~SECRET~~

run be long enough to permit bake-out and adjustment phases, yet allow innage to be of reasonable duration. This required a low charge consumption rate and, consequently, a small total ion current. These conditions were acceptable since previous experience indicated that ion beams of the order of one ma would give isotopic resolution necessary to meet the product purity specification. Low output had the added advantage of increasing the life of the receiver which would be inherently weak due to its small structural size.

These considerations were the basis for specifying a twelve-inch radius, single-arc calutron, enclosed by a correspondingly small liner. The ion exit slit was to be three inches long with a narrow aperture. This equipment was to be operated in a Beta tank from which the magnetic shims would be removed.

Other features believed desirable were the following:

1. A six-inch D.P.I.-500 diffusion pump for evacuating the liner. This feature was to aid in retaining material within the equipment since, when the ion source and receiver were in place, it would cover the remaining opening in the liner.
2. Standard external mechanisms allowing in-and-out and rotational motion of the receiver. The urgency of the project necessitated the use of standard equipment and techniques wherever possible.
3. A charge bottle having a maximum capacity of 30 grams of UCl_4 .

~~SECRET~~

~~SECRET~~

Features allowing the charge bottle to be inserted into, or removed from, the source oven while the unit was in the tank were desired to aid in controlling the uranium sublimation rate during bake-out.

ESTIMATES MADE BY THE Y-12 ISOTOPE RESEARCH AND PRODUCTION DIVISION

The original estimate of product quality and quantity, based on known production rates for existing equipment, anticipated ion currents from proposed equipment, and the expected isotopic distribution in the initial material of 95 per cent U-235, one per cent U-236, and four per cent U-238, stated that 8 to 10 milligrams of uranium would be collected containing 20 to 25 per cent U-236, 20 to 25 per cent U-238, with the balance U-235.

Later, a 100 milligram sample of the U-236 bearing material was received at Y-12, and mass analysis ¹ showed the following isotopic composition:

U-234	0.87%	±0.03
U-235	92.72%	±0.08
U-236	0.67%	±0.03
U-238	5.85%	±0.03

As a result of this assay, a second estimate was prepared which stated that from 4 to 6.8 milligrams of uranium containing 15 to 10 per cent U-236, more than 75 per cent U-235, less than 10 per cent U-238, and about 0.1 per cent U-234 would be collected. The details of this estimate are given in Appendix A.

1. Supplied by R. F. Hibbs, Mass Analysis Laboratory, Y-12.

~~SECRET~~

~~SECRET~~

PRELIMINARY EXPERIMENTAL PROGRAM

The urgency of the U-236 collections indicated that an experimental program should be undertaken, concurrent with the design and fabrication of the 12-inch radius unit, to learn as many of the critical operating characteristics as possible, and to determine additional specifications for the equipment. Principal features investigated were focussing properties, baffle structures in the tank, and receiver design.

The program was carried out in equipment simulating the 12-inch radius unit for uranium but using BiCl_3 charge material. Use of a non-isotopic element expedited determination of the ion beam distribution at the receiver and BiCl_3 had the advantage of eliminating chemical recovery necessary with uranium. From these experiments it was determined that a 12-inch radius calutron was operable at 8-11 kv ion source voltage, and that the electrode system should be approximately half-size Beta 15° geometry for good ion beam focussing. Furthermore, the position and aperture of the 90° baffle were critical and were required to be readily adjustable to reproducible settings. It was indicated that the receiver should have at least two monitoring electrodes in addition to the U-236 collector in order to obtain preliminary focus with minimum contamination of the U-236 pocket.

From studies of the distribution of Bi^+ ions at the receiver, the effects on operations of equipment variations at the 90° baffle and ion source regions were determined. A graph of Bi^+ ion distribution is shown

~~SECRET~~

~~SECRET~~

in Figure 1. Previous studies with bismuth and other elements had indicated the quality of focus to be independent of the element and a function of equipment only. The distribution of the uranium isotopes collected in the U-236 pocket was calculated from the bismuth beam shape yielding the following estimate:

U-234	0.03%
U-235	61.8%
U-236	25.7%
U-238	12.3%

This calculated estimate was verified by performance of the 12-inch radius equipment during test runs with uranium prior to the actual collection runs.

CHARACTERISTICS OF THE 12-INCH RADIUS UNIT USING UCl_4 CHARGE

The special calutron was being designed and built concurrently with the preliminary experimental program according to the general specifications previously listed. Additional specifications were:

1. Locating the terminal box and G bushing so that standard electrical jumpers, and G and M high voltage water leads could be used.
2. Spacing the M and G bushings to make possible the use of standard G adjustment mechanisms.
3. Mounting the ion source and receiver on one face plate.
4. Designing the face plate for mounting on the chemical recovery monorail.
5. Locating the liner door by dowels on the main door.

FIGURE 1. BISMUTH BEAM SHAPE

~~SECRET~~

6. Locating the face plate by dowels on the liner door.
7. Providing the six-inch diffusion pump with a means for rapid cooling.
8. Freeing all liner joints from sharp corners.
9. Providing sufficient distance from arc exit slit to the face plate to assure the operation of the arc in a uniform magnetic field.
10. Providing a leak valve and gas line to the defining slot to permit use of nitrogen arc support gas.

After delivery of the source and liner (Figures 2 and 3), a series of experimental runs were made which led to further modification of the original design. These modifications included:

1. Use of a small, non-retractable charge bottle.
2. Use of a cast-in J heater.
3. Elimination of the six-inch diffusion pump.
4. A movable 90° baffle having an $8\frac{3}{4}^\circ$ aperture.
5. Development of a new receiver design consisting of three defining slots backed by two electrodes and a pocket.

The ion entry slots in the mass 234 and 235 positions were each 0.015 inches by 3 inches with a $\frac{1}{8}$ inch bridge in the center. The 236 mass position slot was 0.025 inches by 3 inches, and could be covered by a shutter. The pocket in the mass 236 position, a cross section of which is shown in Figure 4, was of three-piece construction.

These experimental runs, in addition to indicating improvements to be made in the equipment, afforded an opportunity for making exhaustive

~~SECRET~~

18

FIGURE 2. 12-INCH RADIUS CALUTRON

19

FIGURE 3. 12-INCH RADIUS CALUTRON LINER

20

FIGURE 4. CROSS SECTION OF U-236 RECEIVER

~~SECRET~~

tests on operating techniques, ion beam distribution at the receiver, receiver retention, and chemical recovery efficiency. From data obtained on these exploratory runs, the estimate on product purity calculated from the bismuth beam shape was verified. A feasibility report was submitted to the Atomic Energy Commission and approval was granted to proceed with processing of U-236 bearing material. Details of the collection operations, which yielded 2.6 mg U-236 in a total of 12.4 mg uranium (21 per cent) are given in Appendix K.

CHARGE CHEMISTRY

Concurrent with the operational experiments, work was initiated on the charge preparation and material recovery phases of the program. Although neither problem was new, the scale of operations and the desired high recovery necessitated a development program.

Of the many recovery processes which have been used in Y-12, the HCP² process seemed best suited. Most of its chemistry can be carried out wet, the only exception being the dry transfer of UCl_4 to the charge bottle. Since no high temperature or high pressure equipment was involved, small scale apparatus for the process was of simple design and comparatively inexpensive. Salvage material could be kept to a minimum, and control over the various phases of recovery could be set up simply. The steps of the process are listed below:

-
2. Reaction of uranium oxides with liquid hexachloropropene at atmospheric pressure.

~~SECRET~~

~~SECRET~~

1. Evaporate uranyl nitrate solution to a dry cake.
2. Pick up this cake in concentrated hydrochloric acid.
3. Re-evaporate to a chloride cake.
4. React this cake by refluxing it with HCP.
5. Remove the reaction side products by distillation.
6. Wash and filter the resultant UCl_4 slurry with CCl_4 .
7. Bake the material under reduced pressure at $350^{\circ}C$.
8. Transfer to charge bottle and run in calutron.
9. Wash the unit with dilute nitric acid and water.
10. Evaporate wash solution to a small volume.
11. Extract uranium with diethyl ether.
12. Backwash ether solution with water giving uranyl nitrate.

The original sample, as received by Y-12 from the Oak Ridge National Laboratory, was in the form of uranyl nitrate containing about 12.07 grams of uranium. Two samples were removed from this solution, one containing 0.41 grams of uranium and the second 0.36 grams of uranium, leaving 11.30 grams of uranium available for charge preparation. The first of these two samples was sent to the Mass Analysis Laboratory, and the second to the Isotope Physics Laboratory.

The 11.30 grams of uranium were recycled through the calutron eight times; and at the conclusion of the last recycle, 8.8 grams of uranium remained. The recovery figures calculated from the above data are listed below:

~~SECRET~~

SECRET

A. 8.8 grams of uranium remaining = 77.9 per cent of the original.

Total loss, 2.5 grams = 22.1 per cent.

B. Average recovery per run = 97.34 per cent.

The actual recovery in grams for each individual run is not known as no analysis or accurate weighing of the charge material during recycle operation was possible due to the small quantities involved.

PRODUCT RECOVERY

The recovery of enriched samples of material from the receiver pockets also required development work. After working with various designs, the three piece collector shown in Figure 4 was evolved and found to be most suitable, both from operational and recovery standpoints. This entire graphite assembly was purified by chlorination at high temperature before the collection run. After purification only 4 to 10 parts per million of impurities remained in the graphite as shown by ashing of test samples.

After a collection run, the pocket containing the uranium was broken into small pieces, placed in a platinum dish, and ignited in the presence of excess oxygen at about 850°C. After cooling, the dish was removed from the furnace and the sample put into solution with 10 ml of concentrated nitric acid and a few drops of 30 per cent hydrogen peroxide. The solution was then filtered to remove the small amount of insoluble material. The filtrate was saved and the filter paper was analyzed for uranium. The uranium content of the filter paper was never found to be greater than two

SECRET

~~SECRET~~

micrograms, hence no recovery was deemed necessary.

The nitric acid filtrate was evaporated to dryness and the dry salts redissolved in a saturated solution of ammonium nitrate containing 5 per cent nitric acid. The resulting solution was transferred to a 60 ml separatory funnel and 10 ml of dibutoxytetraethylene glycol were added and mixed by shaking. After the two layers had settled, the aqueous layer was drained off. Ten ml of carbon tetrachloride and 20 ml of distilled water were added to the ether layer in the separatory funnel and then shaken. After the two layers had separated, the fractions were withdrawn into small beakers. The ether layer was analyzed for uranium by fluorescence, and in no case, were more than two micrograms of uranium found.

The aqueous layer, now containing the uranium, was treated with concentrated nitric acid and hydrogen peroxide, destroying all organic material. The solution, approximately 5 ml, was transferred to a small tared platinum vessel. It was then evaporated and ignited to constant weight at 850°C to the form of U_3O_8 . The oxide was then dissolved in a minimum of nitric acid and made up to a volume such that 1 ml of solution would contain 100 micrograms of uranium. A 1 ml sample was removed and sent to the Isotope Physics Laboratory for assay. The remaining volume was reduced, transferred to a glass tube which was then sealed and was ready for shipment.

Because of the small quantities of material involved, only estimates of the efficiency of recovery and the chemical purity of the

~~SECRET~~

~~SECRET~~

sample are possible. The estimates indicate that about 99.8 per cent of the uranium was recovered and that it was about 99.5 per cent pure.

HEALTH PHYSICS

Due to both the toxic effects of uranium, and in this case, the radioactivity as evidenced by Alpha emission, great care was necessary in handling the material. To protect the personnel involved, the Health Physics Department took the following steps:

1. Adequate ventilation systems were provided to remove as much as possible of any airborne contamination.
2. Continuous and complete monitoring of the various work areas was provided.
3. All pipetting was done by mechanical means.
4. Rubber gloves were used throughout by all personnel coming in contact with either the material or contaminated equipment. Regular surgical techniques were used for application and removal of these gloves.
5. Adequate respirators were provided and worn by all personnel in areas where airborne contamination might be present.
6. No food was permitted in areas where active material was used.
7. Monitoring devices were provided for hand checks to be used before smoking, eating, or leaving work.
8. Urine analyses were run on a continuous basis on the personnel directly handling the active material.
9. All work areas were restricted to personnel directly involved.

~~SECRET~~

10. All areas involved were checked and cleaned at frequent and regular intervals.

11. Shoe covers and protective clothing were provided for all personnel in contaminated areas.

12. All protective equipment was properly disposed of and cleaned regularly. This, along with regular daily change of clothing, insured excellent control of potential dangers.

SPECTROGRAPHIC ASSAY

Another problem was that of determining the isotopic composition of the product. Estimates were made as to the product compositions from calutron operational data and were corrected after weighing of the recovered products. This type of estimate is usual, but normally is substantiated or refuted by accurate mass analysis. At the time of this work the Mass Analysis Laboratory had not developed techniques for assaying microgram quantities of uranium. However, the Isotope Physics Group (Spectrographic Laboratory) had developed a method for assaying uranium based on the isotope structure of the $4244 \text{ \AA}$ spectrum line.³ This work had been checked by the group with results from mass analysis instruments and found to be valid.⁴

3. Long, D. R., Smith, D. D., "Observed Differences in the Arc Spectra of U²³⁵ and U²³⁸", CEW-TEC, B-4.380.121.

Clark, F. E., "Spectrographic Assay Results" - CEW-TEC, B-4.380.160.

4. Stukenbroeker, G. L., Adams, S., Burkhardt, L. E., "Isotope Shifts in Uranium Spectra" - Y-20.

Stukenbroeker, G. L., Adams, S., Burkhardt, L. E., "Isotope Shifts in Uranium Spectra" - Physical Review, Vol. 75, No. 1 pp 83-85, 1-1-49.

Smith, D. D., Burkhardt, L. E., - "Mass Analysis of Uranium Isotopes by Emission Spectroscopy" - Y-226.

~~SECRET~~

The method developed and used by this group for the assay of the product samples was as follows:

1. About 20 micrograms of uranium in nitrate solution were absorbed in a filter paper approximately $1\frac{1}{4}$ cm square.
2. The paper was dried, charred, and then ashed in a platinum crucible at about 600°C .
3. About 40 micrograms of the ash was transferred to the crater at the apex of a cone-shaped electrode and sealed with collodion.
4. The sample was arced before a 21 foot Jarrell-Ash, 15,000 lines per inch spectrograph (the instrument was operated in second order at a dispersion of about 4.8 angstroms per millimeter).
5. The arc was operated for about 15 seconds, after which time the contribution to the uranium spectrum was negligible.
6. The plates were calibrated by burning a 20 milligram sample of 40 per cent U_3O_8 in graphite powder for 15 seconds.
7. The resulting spectrograms were densitometered and relative intensities of the three isotopic components obtained. The percentage concentrations were then determined by taking ratios of the isotope component intensity to the total intensity of the three components (U-234 was assumed zero). Corrections were made for background and overlap.

A summary of the assay results is shown in Table I, Appendix K.

~~SECRET~~

CONFIDENTIAL

CONCLUSIONS

As a result of the work described it has been found possible to separate and collect small, highly enriched samples of uranium from a very small quantity of charge material using a special 12" radius calutron.

The recovery of processed material which averages over 97% per cycle, can be accomplished by adapting standard Y-12 procedures.

Small samples containing 100 micrograms of uranium can be assayed with fair accuracy by spectrographic methods.

The Atomic Energy Commission requested 1 mg of U-236 in a total of 6.67 mg of uranium (i.e., 15 per cent) as a product requirement. The six samples collected comprised 2.6 mg of U-236 in a total of 12.4 mg of uranium (i.e., 21 per cent), thus filling the requirements.

~~SECRET~~

APPENDIX A

~~SECRET~~

APPENDIX A

Methods of Estimation of Product Purity

Estimates of anticipated product quality and quantity were required before experimental work with the proposed equipment was undertaken. These estimates were based on the known operational characteristics of both Alpha and Beta equipment, on experience gained in the separations of other isotopes, and on anticipated operating characteristics of the proposed equipment.

The first estimate was based on an expected isotopic distribution in the charge material of 95 per cent U-235, 1 per cent U-238, and the balance U-235.

A later and more reliable estimate was made after mass analysis of a 100 milligram sample of the U-236 bearing material showed it to have the following isotopic composition:

U-234	0.87%
U-235	92.72%
U-236	0.67%
U-238	5.85%

The methods of making this estimate are given, in some detail, below to indicate the difficulties and uncertainties in predicting the performance of untried equipment, and to afford a comparison with later achievements.

I. Estimate of Collection Rate

The ion collection rate expected from the proposed equipment was estimated on the basis of the known performance of standard Beta equipment. With Beta equipment it was known that an average collected U^+ ion current of 85 ma could be obtained from a single arc. A corresponding figure for the proposed equipment was derived by considering those differences between the two types of equipment that appeared likely to modify the collected ion current in more or less predictable ways.

A. Effect of Ion Exit Slit Area

The ion exit slit of the proposed unit was to have an area of approximately one ninth ($1/9$) that of the Beta unit. (Proposed slit: $3/32" \times 3" = 0.281$ sq. in; Beta slit: $3/8" \times 7" = 2.625$ sq. in.) This reduction in slit area was expected to reduce ion output, but not in proportion to the reduction in slit area.

Experience in previous stable isotope operations indicated that the reduction in output would be substantially less than the reduction in slit area; consequently, a factor of five (5) was taken as a more realistic estimate of the resulting ion output reduction.

B. Effect of Beam Trajectory

In the proposed unit the ion beam was to be collected on a 12" radius; whereas, in Beta and Alpha units the beam path radius is 24" and 48", respectively. Moreover, it was known

~~SECRET~~

that the 85 ma average ion current collected in standard Beta operation was reduced to about 53 ma when output from the same source was collected at the Alpha radius. Thus, halving the radius (48" to 24") has the effect of increasing the collected current by a factor of $85/53 = 1.6$. The full implications of this factor are not understood; however, the effect is real and appeared to justify the application of a similar factor to the new equipment in which the radius was to be again halved (24" to 12").

C. Effect of 90° Baffle Opening

A reduction in collected current was expected as a result of reduction in the 90° baffle opening. In the proposed unit this opening was to pass beams of only 4° angular divergence; whereas, the Beta baffle permitted the passage of beams having a divergence of 24°. This six-fold reduction in the angular aperture of the baffle would result, it was estimated, in reducing the collected current by a factor of four (4). This estimate was based on experimentally determined relations between collected current and baffle aperture established in earlier beam studies.

D. Source Voltage

In reducing the ion beam radius from 24" to 12" it was

~~SECRET~~

proposed to increase the magnetic field to near the maximum value attainable, and decrease the source voltage as required to attain the desired radius. This would result in an operating source voltage of 7 to 10 kv as compared with 35 kv normal Beta operation, and was expected to reduce ion collection by an estimated factor of 3 to 5.

Combining the above factors, and taking 85 ma as the ion current from a standard Beta arc, one obtains as the U^+ ion current estimate for the proposed unit:

$$85 \times 1/5 \times 1/4 \times 1/5 \times 1.6 = 1.36 \text{ ma.}$$

Among the factors not determinable at the time this estimate was made, and not taken into account in the above analysis, were: (a) a possible change, due to scaling down of the source unit, in the ratio of U^+ to other ions; and (b) the effect of operating in a homogeneous magnetic field rather than in the normal Beta field into which ion focusing inhomogeneity had been introduced.

II. Estimate of Charge Consumption Rate and Process Efficiency

The charge consumption rate in normal Beta operation was well established as 7 g UCl_4 per hour per arc, or 4.4 g U per hour per arc. Assuming that charge consumption rate is proportional to ion exit slit area, the rate for the new unit would be 1/9 that for a Beta unit, i.e., 707 mg UCl_4 per hour or 490 mg U per hour. If, on the other hand, it

is assumed that charge consumption rate is proportional to ion source output, the new rate would be estimated as 1/5 that for Beta, i.e. 1270 mg UCl_4 per hour or 880 mg U per hour.

The lower charge consumption rate (490 mg U per hour) was taken as the more probable; and with an available charge containing 14 g of uranium, the maximum run length would be

$$14000/490 = 28.5 \text{ hours}$$

With an average collected ion current of 1.4 ma, this gave as the total metered collection per run,

$$28.5 \times 1.4 = 40 \text{ milliamperere hours}$$

One ma-hr of U^+ ions is equivalent to 8.88 mg of uranium; therefore, the maximum total collection per run would be

$$8.88 \times 40 = 355 \text{ mg uranium,}$$

and the maximum process efficiency

$$\frac{355}{14000} \times 100 = 2.56 \text{ per cent.}$$

III. Estimate of Product Quantity

Of possibly 355 mg uranium expected to reach the receiver only 0.67 per cent, or 2.4 mg would be U-236. Not all of this would be received and retained in the collector pocket, however. If one assumes that 30 to 50 per cent of the 2.4 mg of U-236 reaching the receiver enters the pocket, and that 85 per cent of this is retained, the collector would be expected to retain between

~~SECRET~~

$$2.4 \times 0.3 \times 0.85 = 0.61 \text{ mg, and}$$

$$2.4 \times 0.5 \times 0.85 = 1.02 \text{ mg}$$

of U-236.

IV. Estimate of Enhancement

An estimate of the enhancement to be expected in the proposed separation was based on the known average Beta enhancement of about 40 for natural uranium. It was estimated that this enhancement would be modified due to the following causes by the factors indicated:

a. Due to the fact that the critical isotopes (U-235 and U-236) are separated by only one mass unit as contrasted with the three mass units separating the principal isotopes of natural uranium, it was assumed that the enhancement would be decreased by a factor of about 10.

b. Use of an ion beam of $\pm 2^\circ$ angular divergence rather than a total divergence of 24° was expected to increase the enhancement of a factor of about 4.

c. Operation at a 12" rather than a 24" radius should reduce scattering of a factor of 1.25 to 2, and it was assumed that the enhancement would be increased by the same factor.

Thus for the new unit the enhancement should not be less than

$$\frac{40 \times 4 \times 1.25}{10} = 20$$

and might be as great as

~~SECRET~~

~~SECRET~~

$$\frac{40 \times 4 \times 2}{10} = 32$$

For the lower enhancement, the product would be 12 per cent U-236; for the higher, about 17 per cent.

Assuming an average value for the estimated enhancement such that the product contains 15 per cent U-236, one obtains as the total uranium collected in the U-236 pocket something between

$$\frac{0.61}{0.15} = 4.07 \text{ mg}$$

and

$$\frac{1.02}{0.15} = 6.8 \text{ mg}$$

On the basis of these assumptions, the estimated collection would be from 4 to 6.8 mg of uranium containing 15 to 10 per cent U-236, more than 75 per cent U-235, less than 10 per cent U-238, and about 0.1 per cent U-234.

~~SECRET~~

APPENDIX B

~~SECRET~~

~~SECRET~~

APPENDIX B

Preliminary Experimental Program

A preliminary experimental program was conducted concurrent with the design of the equipment to minimize subsequent modification and to determine whether the unit would operate at an accelerating voltage of 8-11 kv, a limitation imposed by magnetic field and ion radius.

One of the main problems at that time was the choice of a suitable charge material which would give results comparable to those expected from the final material, yet sufficiently simple for ready analysis. UCl_4 was undesirable because of its hygroscopic properties and the time involved in its recovery. To simplify ion distribution studies, a nonisotopic element was desirable; BiCl_3 was selected as the charge material best meeting all of these requirements.

In interpreting the BiCl_3 experiments, the basic assumptions were made that the focussing properties of the equipment would be the same for U^+ ions as for Bi^+ ions; and that each of the isotopic ion beams of uranium would have the same properties as the single Bi^+ ion beam. The purity of uranium product to be expected was to be determined by plotting the various uranium beams, each like the Bi^+ beam, in their proper locations along the abscissa. The ordinate (ion current) of each beam was to be proportional to the abundance of the isotope involved, with the measured width of focus being the same for all isotopes. Addition of the various isotopic ordinates at any specific abscissa location (mass

~~SECRET~~

~~SECRET~~

location) would give the total beam strength and the fraction that each isotope contributed to this total.

To simulate the operating characteristics and physical features of the final unit, the preliminary operations were made using a modified Beta M-12 source and receiver. These were mounted so that 12-inch radius operation could be obtained in a Beta tank from which the magnetic shims had been removed. The equipment included a 90° baffle adjustable both as to size and location of aperture, a single arc, ion exit slits of various sizes, standard collimating slots and filaments, and about one-half size Beta electrodes and geometry. The electrode system was supported in the conventional manner. Both zero and 15° electrodes were tested; however, only 15° ion exit slits of various sizes were tried. The source was modified by machining flat the oven and arc casting support brackets, and mounting a flat carbon plate on the front of the unit to support the arc chamber and oven at the upper arc (J-1) position. A copper cylinder with a brazed-on external calrod heater formed the oven into which a charge bottle and standard thermohm were inserted. The arc chamber was modified to permit use of a removable ion exit slit; and a tube was provided for introducing N_2 arc support gas. The vapor baffle was such that the vapor entered the arc chamber through slots at the top and bottom of the rear section.

A standard Beta liner, was so modified as to accomodate a receiver 24" above the center line of the arc; and a framework was provided to

~~SECRET~~

~~SECRET~~

support a movable baffle at the 90° position. A special louvre in the pump-out section of the liner was also provided to prevent passage of ion beams into the tank manifold region.

The receiver, mounted on a $3/4$ " stainless steel rod, consisted of three electrodes behind a single straight 0.015 " x 5 " slot cut in a $1/32$ -inch thick section of graphite. The three electrodes were placed so that the center electrode monitored the central three inches of the ion beam while the other two electrodes monitored the beam entering the one-inch sections at either end of the slot.

The 90° baffle consisted of two stainless steel frames each of which was protected from the ion beam by an attached electrically insulated graphite sheet. These sheets were parallel to the magnetic field and defined the angular divergence of the beam passing the baffle. Each frame was provided with a push rod so that by independent motion of the baffle sections in a radial direction across the ion beam it was possible to vary the location and size of the aperture in the baffle.

During the tests, source problems were given initial attention since most of these affected arc operation and ion beam focus as observed at the receiver. Heat feed-back from arc chamber to oven was reduced by inserting a stainless steel tube between the oven and the carbon support plate. High pressure around the source unit was overcome by sleeving all inside joints of the arc support gas system.

Initial accelerating electrodes were half-size Beta 0° geometry

~~SECRET~~

~~SECRET~~

with a $3/32$ " x 3", 15° ion exit slit. Subsequently, 15° electrodes were used which yielded adequate focus at the receiver. Ion exit slits of many widths were tried with $3/16$ " being found most satisfactory. The G (negative) electrode aperture was lengthened to overcome focussing parallel to the magnetic field; and a skirt was hung from the C (grounded) electrode in front of the lower part of the source front to introduce a grounded surface between source potential and the ion beam.

Vapor flow from charge bottle to the arc chamber through the stainless steel connecting tube was difficult to regulate until the outlet end of the tube was reduced in size by covering it with a stainless steel disc having a $1/8$ " diameter hole in its center. It was found necessary to protect this disc from negative ion bombardment, and the graphite vapor baffle in the arc chamber served this purpose. Changes in the locations of vapor slots in the graphite baffle had no apparent effect on arc operation.

The original push-rod mechanism of the 90° baffle proved inadequate because locations could not be reproduced. To overcome these difficulties, a screw-type mechanism was designed in which the negative angle baffle section was movable relative to the positive angle section to change the size of the aperture; and both sections were movable together to change the location of the aperture. The mechanism is shown in Figure 5.

Provision was made for applying voltages to the insulated graphite

~~SECRET~~

FIGURE 5. 90° BAFFLE MECHANISM

~~SECRET~~

baffle sections; but after a number of tests from which no beneficial effects were observed, the graphite sections were grounded. The baffle opening giving most satisfactory focus was $-2-3/4^{\circ}$ to $+5-1/4^{\circ}$.

Initial studies of ion beam focus were made by monitoring currents at the receiver either by meter or oscilloscope depending on whether scanning of the ion beam was being performed by changing accelerating voltage manually or by saw-tooth modulation. Both of these methods proved unsatisfactory because arc and source ion focus conditions were found to be critically dependent on source potentials. However, during these tests with the three electrode single slot receiver, it was observed that over 90 per cent of the ion beam was received on the central 3" electrode.

To avoid changing accelerating voltage, it was necessary to design a receiver having a number of parallel electrodes spaced to intercept a wide section of the ion beam. Ideally, these parallel electrodes should be of infinitesimal thickness and placed immediately adjacent to each other; current readings should be taken simultaneously from each, and a composite of these readings plotted to give the ion distribution at the receiver.

To approximate the ideal, a receiver was constructed having seven thin graphite electrodes each insulated by mica. The ion beam reached the electrodes through $0.15" \times 4-1/2"$ slots spaced $0.055"$ on centers. After the first test of this receiver, it was found that the $0.040"$

~~SECRET~~

~~SECRET~~

x 4-1/4" separators between the defining slots were warped in the center. To give added strength, 1/8" bridges were left in the center of the slots by cutting each slot in two sections; and several runs were made with this receiver, all of which were satisfactory. A graph of the ion distribution at the receiver, together with calculated uranium ion distributions in their proper locations are shown in Figure 6.

Calculated product composition at the mass 236 position was:

U-238	12.3%
U-236	25.7%
U-235	61.8%
U-234	0.03%

From these experiments, the basic design features were carried over to the 12-inch radius unit for uranium wherever possible; and the ion distribution studies provided a basis for estimating its performance.

~~SECRET~~

FIGURE 6. CALCULATED URANIUM ISOTOPIC DISTRIBUTION

~~SECRET~~

APPENDIX C

~~SECRET~~

~~SECRET~~

APPENDIX C

General Design of the 12-Inch Radius Unit for Uranium

Design of the 12-inch radius unit was started without preliminary experiment, with specifications being defined as the program proceeded. In general, its parts were to be as small and simple as possible to present a minimum of surface area. It was to operate on a 12-inch radius and have one arc, a three-inch long ion exit slit, and a charge bottle with capacity for 30 grams of UCl_4 . To conserve time, drawings of individual parts were sent to the shops immediately upon completion, and a tentative date of April 1, 1948, was set for the delivery of all source parts from the shops.

General specifications were:

1. The unit was to be operated in an unshimmed Beta tank.
2. The liner was to be of the Beta type reduced in size to correspond to the overall dimensions of the unit.
3. A six-inch D.P.I.-500 oil diffusion pump was to be used to evacuate the liner. This was to cover the only opening in the liner when the source and receiver were in place and was to aid in retaining the material within the equipment.
4. The source design was to make use of as many tested features as possible.
5. The receiver mechanism was to be standard and include in-and-out and rotational motion of the support tube.

~~SECRET~~

~~SECRET~~

6. The receiver, the 90° baffle, and the slit system, were not specified pending the results of experimental work.

In addition, the locations of the terminal box and G bushing were to be such that standard electrical jumpers and G and M high voltage water leads could be used. The spacing between the M and G bushings was to be the same as that on an M-12 Beta source which made possible the use of standard G adjustment mechanisms.

Further specifications included:

1. The source and receiver were to be mounted on the same face plate.
2. The M-E door was to be designed for mounting on the chemical recovery monorail.
3. The liner door was to be located by dowels on the main door.
4. The M-E face plate was to be located by dowels on the liner door.
5. The 6-inch diffusion pump was to have provisions for rapid cooling.
6. All joints in the liner were to be free from sharp corners.
7. A leak valve and gas line to the collimating slot were to be provided to permit introduction of an arc support gas.
8. The distance from the ion exit slit to the face plate was to be great enough to assure location of the arc in a uniform magnetic field.

The M-E face plate was the first part to be designed. Since the distance between receiver slots was only 0.051" for 24" beam path

~~SECRET~~

diameter, spacing between source and receiver was critical; thus a single face plate was used for mounting both. As the source was to be small, a single, deep-fluted, Beta-type M bushing was considered sufficient protection against structural and/or electrical failure; and a single bushing was sufficient to support the negative electrode. To conserve space and to minimize the surface area of the unit, an M-12 type G adjustment was adopted with the C electrode being supported from the cylindrical bushing shield. Since only small ion beams were anticipated, the cylindrical bushing shield was not water-cooled; however, provision was made in the face plate for water-cooling if subsequently needed.

The M support structure consisted of an assembly of primarily copper parts hard-soldered together and provided with passages for water-cooling. A tube extended through the M bushing to enclose water and electrical leads, and was provided with a flange to bear against rubber gaskets on the inner end of the M bushing to form a vacuum seal. The flange supported the arc chamber, filament, and oven support members and a fin shield above the M bushing. Two stainless steel tubes extended from the flange away from the face plate and carried a joining stainless steel plate to which the oven could be attached. A rectangular copper structure hard soldered to the ends of the tubes was provided with machined surfaces for locating the filament leads, the arc chamber assembly, and a heat baffle between the oven and arc chamber assemblies. Three point support was provided for the arc chamber.

~~SECRET~~

The drain system was essentially of the Beta type reduced in size to fit the dimensions of the unit. Like plant model Beta M-12's, no fins were used on the anti-drain side of the unit. The #1 and #2 fins were graphite held in place by graphite cap screws and dowels. The remaining three fins were copper and were brazed to the water-cooled fin shield above the M bushing. The hot shield, or "absolute dump" around the M bushing consisted of a segment of a cylinder attached to the fin shield by screws and bolted to the copper flange at the "hot" end of the M bushing. A pair of concentric split rings around the M bushing and secured to the face plate formed the ground termination. The grounded "blister" shield on the drain side of the unit was supported in the conventional M-12 manner.

The high voltage shield on the anti-drain side of the source was a copper plate with a single bend so that it extended horizontally from the face plate end of the cylinder surrounding the M bushing to the heavy copper flange forming the rear of the support structure, and at a 6° angle with the horizontal from the flange to the J front.

In designing the oven and arc chamber systems, careful consideration was given to the details of the 12-inch radius unit in use at the Radiation Laboratory, University of California with the result that a similar arc chamber structure and retractable bottle arrangement were built. By means of a rod extending through the M bushing and a Wilson seal, it was possible to insert or remove the charge bottle from the heat system. The oven was constructed so that a thermohm could be

~~SECRET~~

~~SECRET~~

placed between the oven and the charge bottle. Heat was supplied by a 1000 watt calrod-type heater hard-soldered to the copper oven. A special thermohm designed by the Y-12 group in collaboration with the Leeds & Northrup Company of Philadelphia, Pennsylvania, was built by Leeds & Northrup. The thermohm was approximately a square, one inch on a side, rather than the conventional shape of a 3" x 3/8" rectangle.

The original design provided for the combination-casting type heating system; therefore, no individual arc chamber heaters were included.

The original arc chamber assembly was of three piece graphite construction. The front section contained two unconnected cavities, one below the other. The lower cavity was opposite the charge bottle opening and formed a manifold for the charge vapor, while the upper cavity formed the arc chamber and was long enough to enclose a floating anode. The ion exit slit and the defining slot were cut through the walls of this cavity. The rear section of the arc chamber assembly contained a tapped hole for the end of the charge bottle and a cavity above this hole fell behind the arc chamber and the manifold. A third section of the assembly consisted of a small baffle so placed that the charge vapor was directed through the manifold into the arc chamber. A sketch of this assembly is shown in Figure 7.

The ion exit slits were of the 15° type 3" long and either 3/16", 1/8", or 1/16" wide. The defining slot was 3/8" wide by 5/8" deep and cut for the curved filament but with the taper such as is used in horse

~~SECRET~~

FIGURE 7. SECTION THROUGH ORIGINAL MANIFOLD ASSEMBLY

~~SECRET~~

shoe type slots. The distance from the center of the defining slot to the axis of the ion exit slit was varied with the width of ion exit slit as follows: $3/16$ " slit - $3/32$ " to J zero; $1/8$ " slit - $1/16$ " to J zero; and $1/16$ " slit - $3/64$ " to J zero.

The G and C electrodes were of the 0° and 15° type and were about $1/2$ standard Beta size. The G had a $3/16$ " x $4\ 1/2$ " aperture. Since the G electrode was to be used with but one arc, its base was constructed with 6° taper from either end toward the center so that electron drain to the side shields would be spread. This feature was first used by C. R. McKinney in his U-234 separations.¹

The chlorine side band baffle was mounted from the grounded electrode. Standard types of electrode hangers, reduced in size, were employed.

The C frame was built up from $3/8$ " stainless steel rod and attached to the face plate. The C frame supported a "dust-pan" or lower ground shield and also the short radius side-band baffle and the adjustable 90° baffles. In addition, drain plates for electrons from the G electrode region, were supported from the dust pan and held in place by screws fastened to the blister shield. Two heavy stainless steel channels were welded to the upper section of the C frame and were used as guides for the 90° movable baffle sections.

1. McKinney, C. R. "The Production of U-234 by a Modified Calutron". D-4.250.30.

~~SECRET~~

~~SECRET~~

The 90° baffle consisted of two sections so that the position and size of the aperture could be varied independently. The motion of each section was controlled by a push-rod operated through the M-E door.

Immediately above the C frame, on the M-E door, a small bracket supported a Beta Phillips ion gauge.

The liner was fabricated in a manner similar to that used for the Beta liners and is shown in Figure 3. In order to reduce losses from the system a six-inch stainless steel diffusion pump was connected to the single liner outlet.

A standard Beta E-2 adjustment mechanism was provided, but receiver details were postponed pending the findings of the preliminary experimental program.

~~SECRET~~

APPENDIX D

~~SECRET~~

~~SECRET~~

APPENDIX D

Handling Equipment

Since the 12-inch radius unit, liner, and main door assemblies were departures from standard plant equipment, special handling equipment was required. Wherever possible, existing handling equipment was modified to meet this requirement.

The small liner, when positioned in its main door, formed an assembly which interfered with the "A" frame of the standard Beta transfer truck. Since the 12-inch radius unit face plate was not as wide as the standard Beta face plate, and was to be located in a higher position, it could not be attached to the retractable carriage of the transfer truck.

Modifications of the transfer truck included removal of the retractable carriage and its tracks, and of the "A" frame superstructure without damage to its movable base. A support frame similar to that normally carried by the retractable carriage was then attached to the movable base to carry and properly locate the main door, liner and 12-inch radius face plate during their installation in the tank.

Only minor modifications to monorails, servicing dollies, crane bails, etc., were required for handling the equipment during chemical washing, servicing and storage periods.

~~SECRET~~

~~SECRET~~

APPENDIX E

~~SECRET~~

APPENDIX E

Ion Source Development

The ion source first used in the 12-inch radius unit was a reduced size Beta type and has been described in Appendix C. Design and fabrication proceeded concurrently and as rapidly as possible with the result that by April 1, 1948, basic parts of the ion source were available for testing. The new liner was not available, however, and initial tests were made by mounting the new source parts on a standard Beta M12 face plate and using it with the liner baffle and receiver assembly previously used for the bismuth experiments described in Appendix B.

Initial tests were made using a nitrogen arc to determine whether the unit would operate below 250°C thereby permitting the use of BiCl_3 charge to obtain correlation between the preliminary experimental source and the source designed for uranium. Thermal characteristics of the new source were, however, markedly different from the experimental source as evidenced by heat feed back from the arc region to the charge bottle. It was decided that the new source was inoperable between 120° and 250°C (the operating range for BiCl_3) without extensive modification, and this approach was abandoned.

Uranium impoverished in U-235 (Super-Q) was available and nearly all additional tests were made using UCl_4 prepared from it. These tests included careful examination of the operating characteristics of parts and assemblies, and were particularly concerned with thermal properties as

they controlled sublimation and distribution of the UCl_4 . Also, electrical properties were studied as they affected the arc and the ion beam focussing characteristics. Changes in structure resulted from these tests, and also from the wear imposed by the chemical cleaning of the parts after use.

I. Support Structure Redesign

The M support structure as originally designed included internal water passages vacuum sealed in thirteen places by hard solder and in two places by soft solder. Failures of these hard soldered seals due to repeated acid washes made frequent repairs necessary and led to a new support structure design. In the new design, copper tubing was brazed in grooves machined in the assembly. The ends of the tubes were connected to inlet and outlet tubes with copper sleeves and hard solder. This structure proved to be entirely satisfactory and is shown in Figure 8.

II. Vapor Control

Testing of the source with uranium quickly brought out the fact that, although the design used scaled-down, Beta-type parts and tested principles wherever possible, the operability of the heat system with UCl_4 was unsatisfactory due to erratic vapor flow, charge condensation in the manifold, and abnormally high arc strike temperatures.

In the original assembly, no means were provided for independent heating of the arc and oven systems, nor of effectively preventing heat transfer between them. This was in accordance with existing Beta practice in which a so-called combination casting surrounded the charge bottle with the heat-

60

FIGURE 8. ION SOURCE SUPPORT STRUCTURE

ing elements being placed between the two assemblies. Attempts to adapt this method to the 12-inch radius unit were finally abandoned because the heat losses of the arc assembly were more than the heat system could overcome without excessive heating of the charge bottle.

The trend of experiments, therefore, was to modify parts so that the highest temperature would exist in the arc chamber region rather than at the charge bottle. This approach was finally abandoned for separate arc casting heaters independently controlled, and with the entire arc chamber assembly heat shielded from the oven.

Vapor flow, which in early experiments was very irregular, was improved by increasing the area of the opening from the bottle. The charge bottle shape was changed so that the outlet would not be plugged with UCl_4 ; and so that during operation, the surface area of the charge would not vary appreciably.

A. Oven. One of the various ovens tested is shown in Figure 9. The original premise was that all the heat necessary for both the oven and arc chamber regions could be supplied from one source; but none provided adequate heat for the arc region without overheating the charge bottle. Consequently, the oven was used to heat the charge bottle only.

The final design (Figure 10) used was a copper enclosure, having a brazed-on external calrod heater, surrounded with several layers of stainless steel heat shielding. One difficulty encountered with this oven during operations using U-236 bearing material was hold-up of sufficient material by its surfaces to cause it to be continuously radioactive,

62

FIGURE 9. OVEN AND MANIFOLD ASSEMBLY WITH CHARGE BOTTLE

63

FIGURE 10. FINAL OVEN AND MANIFOLD ASSEMBLY WITH CHARGE BOTTLE

~~SECRET~~

although not dangerously so.

B. Arc casting. First specification for the source called for a single heater casting for both the oven and arc regions, anticipating that the arc would provide enough additional heat to maintain the arc region parts at temperatures above those of the charge bottle and thereby avoid condensation of the UCl_4 . Various changes in the ovens and in the arc region graphite parts were made to accomplish this end with only partial success.

Most of these changes attempted to reduce the volume of graphite in the arc region, the area from which heat could be radiated, and the volume of graphite through which heat could be conducted. Other changes sought to increase the area of heated casting exposed to these graphite parts, and/or provide protrusions on the casting which could be inserted into recesses in the graphite parts. Condensation of UCl_4 , to some degree, accompanied the use of all of these variations.

Upon concluding that no single heater casting would provide sufficient arc region heat without either excessively heating the charge bottle or extensive modification and/or shielding of arc region parts, two types of separate arc casting heaters were tested. The first of these was inserted from the front and the second from the rear. The latter proved unsatisfactory as it could be used as an oven for the vapor manifold and at the same time have surfaces recessed in the graphite front.

During these experiments, the manifold section of the arc assembly had been reduced in volume and was formed by two pieces of graphite

~~SECRET~~

(Figure 11). One piece contained the opening for the charge bottle and a cavity. The second piece covered the cavity, contained a vapor slot, and formed the back of the arc chamber. The piece containing the cavity, or manifold, was exposed to a heater on all sides except that facing the arc chamber. The second piece which formed the back of the arc chamber was exposed on its edges to the heater, but was primarily heated by the arc, negative ion bombardment and radiation from the first section. No charge condensation was ever observed in this assembly.

In the final design, the arc region assembly was insulated from the oven assembly by stainless steel heat shielding.

C. Charge bottle. Originally the charge bottle was cylindrical, loaded from one end over which a cap was placed, and threaded at the other end for engagement with the vapor manifold. These features allowed the bottle to be held on the end of a movable rod for insertion in the oven after outgassing the unit. This feature subsequently was found unnecessary.

The cavity of this bottle was cylindrical with its axis horizontal and it was found quickly the charge as sublimed from the bottle left a surface of irregular but generally increasing area. In effort to overcome this difficulty, a stainless steel screen basket was placed in the bottle to receive the charge and hold it away from the vapor exit. Changes in charge surface during sublimation still persisted, however.

Irregular vapor flow was overcome by increasing the diameter of the vapor exit from 1/8" to 5/32" and later to 7/32". The accompanying re-

66

FIGURE II. SECTION THROUGH FINAL MANIFOLD ASSEMBLY

[REDACTED]

duction in vapor pressure in the bottle made possible striking of an arc at temperatures from 10° to 20°C lower than those first experienced.

The early runs with uranium indicated that charge would not be wasted during initial outgassing with N_2 ; consequently the retractable feature of the bottle was not needed. The bottle was redesigned so that it could be loaded from the bottom rather than the end, and so that it needed only to be inserted into the manifold rather than threaded. The charge cavity was of uniform diameter; and as the charge sublimed, no marked change in surface area was encountered.

D. Temperature Regulation. Temperature regulation of the H oven was obtained in the usual fashion with a thermohm and micromax. Due to the small dimensions, a special thermohm was designed in which the element was within a thin square section ($1\frac{1}{4}" \times 1\frac{1}{4}" \times \frac{1}{8}"$) which could be inserted between the H oven and the charge bottle.

Temperature control was extremely sensitive, thus heater current settings were adjusted frequently during operation to obtain adequate bottle temperature.

Initial use of the arc casting heater included its control from the micromax; but it was found that the loss of heat during a cooling cycle, while having no particularly deleterious effect at the charge bottle, resulted in charge condensation in the manifold. This was overcome by providing a separate manually controlled power supply for the arc casting heaters.

[REDACTED]

III. Electrode Region

The development of the 12-inch radius source included studies of both its thermal and electrical properties; and, for the most part, urgency required that these be studied simultaneously. In the final analysis, the experimental data which determined whether a desired thermal property had been attained were the properties of the ion beam produced by the source and observed at the receiver. These data were unavoidably affected by the types of electrodes used and the characteristics of the UCl_4 arc which existed. When ion resolution at the receiver was attained, basic development ceased and attempts at improvement began. Thus, during early experiments, attention fluctuated between thermal and electrical properties until a combination was found which would consistently yield a focussed ion beam.

The quality of focus attained was measured by observing the peak ion current at one receiver electrode and comparing this with the current observed one mass unit higher. If the ratio of these currents was greater than 30 to 1, satisfactory resolution was considered to have been attained. When such an optimum was reached it was usually found that the values of negative electrode voltage, arc voltage and arc current were very critical, and that a small change in any one would result in loss of resolution. Consequently, operation of the unit required almost constant attention and vernier type control.

Steadiness of ion beam and arc, and absence of unwanted drains and

oscillations required consideration of the general factors influencing quality of focus. These included the ease and reproducibility of assembly of components, the constancy of position of electrodes during the course of the runs and the geometrical arrangement of the electrodes. Also, the assembly was to assist in keeping $\bar{U}$ drains and charge consumption rates to a minimum at maximum ion beam output.

Specific attention was given to:

1. Cross-section area of the arc chamber.
2. Position and type of defining slot.
3. Position and utility of a grounded or floating anode.
4. Length, width, and type of ion exit slit.
5. Location and type of negative electrode.
6. Location and type of grounded electrode.

The following table shows the various combinations arc of chamber assemblies used.

Table 1Arc Chamber Assemblies Tested

<u>Def. Slot</u>	<u>Depth</u>	<u>Width</u>	<u>Ion Exit</u> <u>Slit-Width</u>	<u>Type</u>	<u>Anode Type</u>	<u>Results</u>
1. Curved	3/8"	5/8"	3/32"	15°	Grounded	No focus
2. Curved	3/8"	5/8"	3/16"	15°	Grounded	No focus
3. Curved	3/8"	5/8"	3/16"	15°	Floating	No focus
4. Curved	21/32"	1-1/4"	3/16"	0°	Grounded	No focus
5. Curved and Straight	21/32"	1-1/4"	3/16"	15°	Grounded	20:1 Ratio
6. Straight	21/32"	1-1/4"	3/16"	15°	Floating	no focus
7. Curved	21/32"	1-1/4"	3/32"	15°	Grounded	30:1 Ratio 300 μ a peak
8. Curved	21/32"	1-1/4"	1/8"	15°	Grounded	30:1 Ratio 400 μ a peak
9. Curved	21/32"	1-1/4"	1/16"	15°	Grounded	?

~~SECRET~~

The arc chambers used in #1 through #3 were about 1/2 size Beta shallow arc chambers. The boxes used on the later tests, #4 through #9, were the same type as those used on the bismuth runs in the preliminary experimental program.

Several experimental runs were made with each of the types which gave satisfactory focus in order to determine outputs and charge consumption rates. It was found that as the ion exit slit width was increased the ion output increased markedly and an acceptable focus was increasingly difficult to obtain; and that as slit width was decreased, an acceptable focus was easily obtained but at markedly lower output. Consequently with the wide ion exit slit, the operator struggled to obtain a satisfactory ratio, and with the narrow slit, to obtain adequate output. The net effect was the appearance of a minimum charge consumption rate at about 1/8" ion exit slit width, with which it became relatively easy to obtain both adequate focus and ion output. The final type used was that described in #8 in the above table. Its choice was dictated by consideration of the charge consumption rate, focus, and output obtained.

[REDACTED]

APPENDIX F

[REDACTED]

~~SECRET~~

APPENDIX F

Receiver Development and Beam Studies

Initial consideration of the receiver problem indicated the need for preventing contamination of the U-236 collector during startup and receiver disassembly, reception of probably 20 milliamperes hours of ion current, and a structure whose size and tolerances were such that it could be fabricated from graphite.

Design work was not started on the receiver until preliminary experiments with non-isotopic elements had resulted in the determination of a characteristic ion distribution at the receiver.

I. Monitoring Receivers

Multiple slot monitoring devices and fixed accelerating voltage were generally employed to analyze the beam, since, when using voltage scanning, the ion distribution changed.

The seven slot monitoring receiver used in the preliminary experiments with bismuth was unsatisfactory because material deposited on the mica insulating wafers, shorted the graphite probes together, and necessitated the disassembly of the unit and replacement of the mica.

The next monitoring receiver consisted of five parallel 0.010 in. tantalum wire electrodes, spaced 0.051 in. apart on centers. Each wire was spring loaded to prevent sagging. Support for the wires was constructed from "Microy" blocks, clearance drilled for the wires and mounted in a carbon box, which was in turn mounted on a receiver face

having a $4\frac{1}{2}$ in. x $\frac{1}{2}$ in. opening in front of the wires. With this device, it appeared that focus improved steadily as operating techniques improved. Finally with peak ion beams of about 200 microamperes, negative current was obtained to the electrode one mass unit above that reading the peak current. This phenomenon (observed subsequently with some other receivers) was presumably due to secondary electron emission and made evaluation of ion distribution impossible.

In order to check these observations further, a single 0.015 in. x $4\frac{1}{2}$ in x $1/32$ in. slot was provided on the same receiver $1\frac{1}{2}$ in. above the highest wire. A graphite electrode was placed behind the slot for monitoring. Observations made during operation indicated that it was not possible to move the beam from the wires to the slot without destroying the focus. However, the focus could be restored by small adjustments of the arc conditions, G voltage, slits and baffle. Since adjustments were required, it was felt that the results were qualitative only. Current monitored at the electrode indicated that the focus was not as sharp as evidenced by monitoring at the wires alone. Due to these difficulties and erosion of the wires, this type of monitoring receiver was abandoned.

The next monitoring receiver had two graphite electrodes spaced 0.036 in. apart; a face plate having a $4\frac{1}{2}$ in. x 0.066 in. slot shielded the electrodes so that only 0.015 in. of each was exposed. A third electrode was provided behind and between the other two. Since these electrodes did not receive equivalent fractions of the incident ion

~~SECRET~~

beam, a new face plate having three 0.015 in. x 4 in. x 1/16 in. slots spaced 0.055 in. on centers was substituted. Small spacers were positioned centrally in the slots to maintain their alignment. All three electrodes were similar to those used with the previous receiver. This receiver was satisfactory.

Using charge material enriched in U-238 (Super Q) peak currents of 700 microamperes were monitored, and average values between 300 and 500 microamperes were expected in the production runs. The focus of the nonisotopic beam improved and ratios between the monitored peak and the monitored current one mass unit above the peak were 35 to 1 or better. The results indicated that the ion distribution with uranium was the same as that obtained with bismuth (Figure 1).

II. Collection Receivers

Theoretically, monitoring could be done using either the U-235 peak or the U-238 peak during a U-236 collection run. However, isotopic assays showed the U-235 to be approximately 25 times as abundant as the U-238, which meant that the U-238 ion current peak would average less than 20 microamperes. Considerable difficulty had been encountered in monitoring small ion currents; therefore, the U-235 was selected as the beam to be used for monitoring.

A. Collection Receiver No. 1

The first collection type receiver had three entrance slots, one pocket and a shutter. The slots were spaced 0.051 in. on centers in

~~SECRET~~

a 3/16 in. thick graphite plate. Each slot was 3 in. long, 0.010 in. wide and a 1/8 in. bridge was left in the center of each of the two lower slots to reduce warpage during operation.

To reduce the ratio of the depth of each slot to its width, the graphite plate was reduced to 1/16 in. thickness in the region including the slots. A graphite pocket for the U-236 was placed behind the top slot, and insulated graphite probes were placed behind each of the two lower slots.

Two Lava Studs fastened to the graphite plate served to support the probes and pocket and insulated them from ground. Lava washers insulated them from each other and stainless steel nuts threaded on the studs clamped the entire assembly in place. Clearance between the probes, pocket and ground face were extremely small, 0.007 in. - 0.003 in.

A Standard Beta #2 receiver mechanism was used to provide in and out and rotational adjustment. A graphite shutter for the U-236 slot was provided; and set screws prevented lowering the shutter over the other two slots. During startup the shutter closed the U-236 slot, and the U-235 ion beam was peaked on the lowest slot. The ratio of ion currents at the 235 and 236 mass positions was then checked by monitoring on the lowest and middle electrodes. When the ratio was high enough (preferably above 30), the beam was raised one slug, the U-235 beam peaked on the middle slot, and the shutter raised to allow the U-236 ions to enter the pocket.

During the second week of June, 1948, the unit was charged with

~~SECRET~~

~~SECRET~~

enriched U-238 and a trial run was made. The receiver was an immediate source of trouble because of the effect of shutter on the beam. When the shutter was closed, the monitored currents on the middle and bottom slots were reduced by half and severe oscillations of the beam were observed. Peak current on this run was 300 microamperes. In subsequent tests, the shutter was removed and the receiver performed satisfactorily.

The receiver finally used was a modification of this general type.

B. Collection Receiver #2

A second type receiver was built to collect in three mass positions. Collection of all ions in pockets was desired to reduce probe to ground and probe to probe shorts, and thereby increase the useful life of the receiver.

The receiver, (Figure 12) was constructed from a solid block of graphite. Three openings were cut in the block; these corresponded to the slots of the ground face of the receiver previously described. Each opening was about $\frac{1}{2}$ in. deep and formed a pocket along the bottom of which an insulated 0.050 in. tantalum wire was placed for monitoring.

This receiver was difficult to assemble since the walls between the pockets were quite thin and the monitoring wires were difficult to insulate from the graphite. Small ceramic beads were placed on the ends of the wires and then placed in the graphite block to serve as alignment bushings. Because of the close spacing of the slots, the top and bottom wires could not be placed directly behind their

78

FIGURE 12. COLLECTION RECEIVER NO.2

~~SECRET~~

respective slots.

This type of receiver was abandoned because monitored currents varied from pocket to pocket as a result of the asymmetry of the electrode arrangement.

No further attempt was made to provide individual pockets at each mass position, and the receiver described generally in section A was finally used. The design adopted involved modifications of nearly every part as the results of the experimental runs indicated the need.

III. Receiver Defining Slot Experiments

Primary emphasis of all the experimental work was placed on improving the performance of the unit toward the goal of obtaining one milligram of U-236 in each run. To accomplish this required an overall process efficiency of at least two per cent. Charge consumption averaged about one gm UCl_4 per hour and innage per run averaged about 10 hours. Hence, ion currents at the receiver of at least 11 microamperes of U-236, and 1300 microamperes of U-235 were required to meet the goal. However, attempts to operate at these currents resulted in receiver failure before charge exhaustion, and subsequent efforts were directed to determining optimum receiver defining slot widths, and maximum feasible collection.

A new receiver face plate was built in which the width of each defining slot was changed from 0.010 in. to 0.020 in. The 0.051 in. dimension between centers of slots was retained.

~~SECRET~~

~~SECRET~~

This receiver was successful in allowing a larger amount of material to be collected but also permitted entrance of more impurities to the pocket. Monitored currents were usually about one milliamperere and often were as high as 1.5 millamperes. Beams of this intensity caused rapid erosion of the small graphite spacers and bridges of the receiver face plate, and damaged the electrodes behind the face plate, eroding holes through their thin sections. To overcome these difficulties a face plate having 0.010 in. slots in the U-234 and U-235 mass positions, but a 0.020 in. slot in the 236 mass position was used. This receiver did not fail during operation; but because the monitored U-235 ion current was decreased, the tendency on the part of the operator was to overheat the source unit.

It appeared that the U-234 and U-235 slots, since both were to be used for monitoring the U-235 beam, should be less than 0.030 in. to prevent receiver failure, but greater than 0.010 in. for satisfactory monitoring. The U-236 slot on the other hand, should be as wide as possible, consistent with the desired enrichment, to allow collection of the greatest amount of material.

To check enrichment factors, several isotopically different uranium charge materials were used. The mass analyses of these are shown in Table 1.

~~SECRET~~

~~SECRET~~

TABLE 1

Mass Analysis of Uranium Charge Materials

<u>Isotope</u>	<u>Super Q</u>	<u>1300 Assay</u>	<u>1900 Assay</u>
U-234	negligible	0.8%	0.8%
U-235	0.04%	29.2%	82.0%
U-238	99.96%	70.0%	17.2%

Using Super Q in the 12 in. radius calutron, resulted in the determination of a characteristic resolved ion beam distribution. This intensity distribution was determined using the multiple slot monitoring device described in Section I of this Appendix. The distribution was similar to that obtained using bismuth.

It was assumed that each of the resolved isotopic beams would have this type distribution and that the total ion intensity of each would be in proportion to its natural abundance. Using Super Q and a three-slot receiver having 0.010 in. slot widths, it was possible to define the resolution in terms of the three ion currents observed when the beam was monitored on the center (235) slot. Since the slots were spaced one mass unit apart, the ion current through the highest (236) slot measured the U-235 contamination in the U-236 pocket, the ion current through the central slot was in proportion to the anticipated U-236 current, and the ion current through the lower (234) slot was indicative of the amount of U-238 contamination in the U-236 pocket.

~~SECRET~~

~~SECRET~~

The ratio of the peak current through the central slot (U-235) to that through the highest slot (for U-236) was maximized and used as the criterion of acceptable resolution.

Using either of the recycle charge materials, the resolved U-238 ion beam was first peaked on the lowest slot and a high energy side ratio of the peak current to that one mass unit higher was determined. The U-235 ion beam was assumed to have the same high energy side ratio, but this could not be determined directly, since the current one mass unit above the U-235 peak included many U-238 ions. This was because the ion intensity on the low energy side was several times that on the high energy side due to the asymmetrical ion distribution at the receiver.

Thus, knowing the peak isotopic ion currents, the high energy side ratio and the ion current at the U-236 pocket, it was possible to predict the assay of material in the U-236 pocket.

For example, using the 1300 recycle material and assuming U-235 and U-238 peak ion currents of 300 and 720 microamperes, respectively, and a high energy side ratio of 50 to 1, the U-235 ion current in the 236 position would be 6 microamperes. Assuming an average monitored ion current at the 236 position of 12 microamperes, then 6 microamperes would be U-235 ion current and 6 microamperes would be 238 ion current, which would result in equal amounts of U-235 and U-238 in the U-236 pocket.

Application of this method required stable operation with a minimum of sparking. The U-238 peak ion current and high energy side ratio

~~SECRET~~

were checked at 20 minute intervals with the U-236 pocket shutter closed. The U-235 was monitored in the middle slot during reception. Esterline Angus recorders were placed in the monitoring circuits so that continuous traces of the three ion currents were recorded.

The results using 1300 recycle material are shown in Table 2.

TABLE 2

Results of 1300 Recycle Runs Pocket Assays
(0.010" - 0.010" - 0.010" Slot Receiver)

	<u>Predicted</u>	<u>Actual</u>
Run #1	235 = 22%	235 = 32%
	238 = 78%	235 = 68%
Quantity	0.712 milligrams	0.750 milligrams
Run #2	235 = 30%	235 = 29.5%
	238 = 70%	238 = 70.5%
Quantity	1.045 milligrams	1.1 milligrams

Before the 1900 material was run, the U-236 mass position defining slot was enlarged to 0.030 in. to increase the collection rate. The assembly of this receiver required a new U-235 position electrode insulated from the 236 pocket by mica. The defining slots at the U-235 and U-234 positions were 0.015 in. wide.

The 1900 recycle runs, results of which are shown in Table 3, were not conclusive since the operation was not stable. However, it is significant that the quantity of material collected in all cases was greater than that predicted. If it is assumed that the extra material collected has the same isotopic composition as the starting material, the

revised estimates are obtained which agree more closely with the actual results.

TABLE 3

Results of 1900 Recycle Runs
Pocket Assays

(0.018 in. - 0.018 in. - 0.030 in. Slot Receiver)

	<u>Predicted</u>	<u>Actual</u>	<u>Revised Estimate</u>
#1	235 = 62.5%	235 = 80%	235 = 75%
	238 = 37.5%	238 = 20%	238 = 25%
	.58 mg		1.69 mg
#2	235 = 43	235 = 80%	235 = 68%
	238 = 57	238 = 20%	238 = 32%
	.595 mg		1.69 mg

The amount of extra material was found to be proportional to the U-236 receiver slot width as shown in Figure 13.

To minimize the possibilities of overheating the source unit with resulting poor resolution, a receiver with a 0.015 in. defining slot in all three mass positions was tried. Operationally, this defining face was the most satisfactory that had been used. However, with this face plate, collection of the desired one milligram of U-236 during an average length run was not possible.

To overcome this objection, the U-236 slot was increased to

85

85

FIGURE 13, RECEIVER CONTAMINATION AS A FUNCTION OF SLOT WIDTH

~~SECRET~~

0.025 in; and to reduce the U-235 contamination the center to center spacing between the 235 and 236 mass position defining slots was increased from 0.051 in. to 0.054 in. This defining slot arrangement was tested and used for the production runs.

IV. Side Band Shielding

Shields were attached to the receiver to prevent contamination due to sidebands. Patterns on the receiver defining face plate indicated that sidebands were being deflected onto it from the top of the liner. The UCl_3^+ and UCl_4^+ sidebands struck the liner several inches in front of the receiver; the UCl^+ beam struck the defining face plate above the slots; and the UCl_2^+ beam struck other parts back of the receiver. Two horizontal canopy shields were provided as shown in Figure 4. The upper canopy shield projected 5 in. in front of the receiver defining face and the lower one projected 1 in.

V. Shutters

Since contamination of the U-236 pocket during startup could not be tolerated, a shutter was required. A modification tried consisted of a graphite strip positioned behind the receiver face plate and movable in a plane parallel to it. The lower edge of the shutter was thin so that it could be positioned between the 236 pocket opening and the corresponding face plate defining slot.

With this type of shutter the only disadvantage appeared to be a build-up of material in the 236 entrance slot. This was subsequently

~~SECRET~~

~~SECRET~~

overcome by improving the face plate design and placing the shutter in front of it.

Since the earliest design of external shutters was somewhat cumbersome and caused beam oscillations probably resulting from periodic buildup of charge on the poorly grounded shutter, the face plate was redesigned so that the shutter could be thin and slide vertically along it. The contact between the face plate and back of the shutter provided improved grounding of the shutter, and in operation no deleterious effects were encountered.

VI. U-236 Pocket

In an early modification, the U-236 pocket and its removable cover were changed to prevent loss of material through openings at the bottom of the pocket when the cover did not fit well. The pocket walls were formed so that the cover could be inset and thus eliminate such losses.

As a guard against possible contamination of the collected material from beams passing through the lower defining slots, a grounded receiver pocket was tried. It consisted of three pieces - a forward section that was mounted on the defining face, a sheet of insulating mica, and an insulated back cover plate that served as a monitoring probe. Erosion patterns in the collection pockets had indicated that nearly all the 236 mass beam would strike this plate. However, it was found that the grounded forward section accumulated some uranium so that it could not be discarded as intended. The volume of graphite involved in chemistry together with fabrication and assembly difficulties led to abandonment

~~SECRET~~

~~SECRET~~

of this collector.

The last change in design was to provide a pocket insert which was set back from the pocket opening. This minimized the deposition of unwanted material on the insert; and in addition, the overall design provided for removal of the insert without complete disassembly of the receiver. With this type of collector (Figure 14) the quantities of uranium recovered approximated the monitored reception.

Throughout the entire experimental program, serious consideration had been given the 90 degree baffle opening and its position relative to the source-receiver plane. Since both were controlled variables, optimum settings could be made at any time during operation.

Geometric studies of beam trajectories at the inception of the project indicated that a maximum of 2 degrees of angular divergence could be tolerated at the 90° position.

In these studies, it had been found that with openings greater than 4° , masses at the higher end of the periodic chart separated by only one mass unit would not be resolved 180° from the source on a 12 in. radius. As expected, however, larger outputs were obtained at greater baffle openings.

Despite the early geometric studies, it was found subsequently that the quality of focus was not destroyed by a total baffle opening up to $8\frac{1}{2}^\circ$ (1.75 in.). Invariably, the baffle aperture was run in the positive angles with the negative edge located near the zero degree position. The

~~SECRET~~

89

FIGURE 14. U-236 RECEIVER (EXPLODED VIEW)

defining face of the receiver was run behind the source-receiver plane, or toward the face plate, approximately $1\frac{1}{4}$ in.

Since an $8\frac{1}{2}^\circ$ baffle opening was determined by experiment to be an optimum size, the baffle sections were locked at this opening. Eventually the baffle was made from a single piece of graphite having a fixed $8\frac{1}{2}^\circ$ aperture, and being adjustable only in its radial position.

One attempt was made to fix the 90° baffle with no adjustment and eliminate an additional operational variable. This run was unsuccessful in that sharp focus could not be obtained. Close inspection of the 45° and 90° baffles at termination showed that an intense section of beam had been intercepted by the short radius side of both baffles. Thereafter, the adjustment feature was retained.

~~SECRET~~

APPENDIX G

~~SECRET~~

~~SECRET~~

APPENDIX G

Charge Chemistry and Recycle Operations

The primary problems in charge preparation and recycling were to determine the applicability of standard Y-12 processes for large batches of uranium to the very small quantities to be used in the U-236 separations, and to assure the high material recovery required.

Of the many recovery processes which have been used in Y-12, the HCP¹ process seemed adaptable to these problems because most of its chemistry is carried out wet--the only exception being the final transfer of UCl_4 to the charge bottle. Since no high temperature or high pressure equipment was involved, small scale apparatus for the process was of simple design and comparatively inexpensive. Salvage material could be kept at a minimum, and control over the various recovery phases could be set up simply.

I. HCP Process Development

Details of the HCP process as developed for the production of kilogram quantities of UCl_4 for plant production are given in another report.² Since there had been no previous experience in the use of this method for processing small quantities, an experimental program was undertaken to establish the feasibility of applying the process to small scale production.

-
1. Reaction of uranium oxides with liquid hexachloropropene at atmospheric pressure.
 2. Adams, R. H., R. S. Lowrie, K. B. Brown, "Closed Cycle Beta Process" Y-12, September 24, 1947.

~~SECRET~~

~~SECRET~~

A series of individual material balance runs were made with normal uranium charge material in conjunction with test runs on the twelve-inch radius calutron. Prime considerations during the development work were safety of personnel, prevention of losses, and improvement of process procedures, techniques, and equipment.

In the final analysis, the only variations in charge chemistry developed during the U-236 collections resulted from the small amount of material involved. This necessitated the designing of small equipment and minimizing of both loss and holdup of material. In addition, nitric acid was used in place of aluminum nitrate for initially cleaning calutron parts since virtually quantitative removal of the uranium was required. Also, final degassing of UCl_4 was carried out in a sealed fritted glass filter instead of a charge bottle. The remaining steps in the recycle operation were unchanged.

II. Radiation Hazard

Because of the high Alpha activity of the charge material, it was necessary to take precautions against hazards to personnel. Safety procedures, as formulated by the Y-12 Health Physics Group, are given in Appendix J.

III. Process Description

In general, the HCP process as adapted for the U-236 collections included removal of the uranium bearing material from the equipment with dilute nitric acid and water. This solution was evaporated to a small volume, pH and specific gravity adjusted, and the uranium extracted with

~~SECRET~~

~~SECRET~~

diethyl ether. Backwashing the ether solution with distilled water gave a solution of uranyl nitrate.

Recycling continued by evaporating the uranyl nitrate solution to a dry cake, dissolving this cake in concentrated hydrochloric acid, re-evaporating to a chloride cake, and refluxing this cake with HCP. The side products of this reaction, chiefly trichloroacrylyl chloride ($\text{CCl}_2 = \text{CCl} - \text{COCl}$), were distilled off and the resulting slurry of UCl_4 was filtered and washed with CCl_4 in a fritted glass filter. This filter was sealed on the open end and inserted into a bakeout apparatus. Organic materials were removed by baking at 350°C and 30 microns pressure. The dry UCl_4 was then transferred to a charge bottle and inserted in the calutron.

Various salvage operations were carried out on carbon parts from the source unit, insoluble residues from the wash operations, and salvage solutions from other recycle operations. These salvage items were combined with the bulk of the material before the ether extraction step.

A. Calutron Run

Material losses occurring during run time in the calutron have never been completely evaluated. However, the following precautions were observed to minimize possible losses:

1. All direct handling of charge bottles and contaminated equipment was done by chemistry personnel.
2. Operations personnel cooperated in loss prevention by

~~SECRET~~

careful operation of heaters, especially during start-up.

3. The liquid nitrogen trap was removed at the termination of each run and the condensate removed for subsequent recovery.

B. Unit Wash

Upon termination of a collection run, the receiver pocket was removed from the unit as soon as possible, wrapped in Kodapak, and sent to the purification laboratory. Chemical purification of the product is discussed in Appendix H.

The remaining parts were washed to remove uranium for subsequent recovery. To minimize losses during this step, the units were dismantled, piece by piece, over large (24 in. x 28 in.) porcelain trays. To obtain a solution of high uranium concentration, the parts most heavily contaminated were rinsed with dilute nitric acid along with, and inside, the body of the liner since it was the largest single piece of equipment to be cleaned and contained more material than any other single part. The resulting concentrated wash solution was processed separately from other salvage solutions.

The once-rinsed parts were returned to the porcelain pans and were cleaned along with less contaminated parts, first with nitric acid, then twice with water. Parts were scrubbed with brushes fabricated completely from stainless steel. The dilute solution from this operation usually amounting to six to eight gallons, was collected for subsequent uranium

~~SECRET~~

recovery. Parts were dried with common cleansing tissues which were later salvaged.

As a test for complete recovery and from health hazard considerations, Alpha counters were used to check for the presence of uranium before releasing source parts for re-assembly. If uranium was detected, the parts were rewashed. Heater assemblies frequently required leaching in concentrated acid before cleansing was complete. In a few cases even this treatment was insufficient and the contaminated parts were removed from the cycle.

C. Uranium Extraction

The concentrated uranium-bearing wash solution (about one liter after addition of the wash from the charge bottle proper) was boiled for one hour with the frequent addition of small amounts of hydrogen peroxide in order to oxidize some insolubles; and was then filtered through #40 paper in a large fluted funnel. The specific gravity of the filtrate was built up to 1.4 with cupric nitrate and the pH was adjusted to 1.6 with calcium hydroxide. These steps were necessary for achieving maximum recovery of uranium and minimizing impurities. Uranium extraction from the concentrated solution was then effected by shaking 750 ml portions with diethyl ether. The discarded material consistently showed less than 1 ppm uranium by fluorometric analysis. The uranyl nitrate, backwashed from the ether with separate portions of distilled water, was ready to proceed to cake formation and chlorination while the spent ether was salvaged for reuse.

The dilute wash solution was filtered and placed in an evaporator

~~SECRET~~

~~SECRET~~

containing a tantalum steam coil heater and fitted with a spiral condenser. The condensate was later re-evaporated and the distillate, when analyzed, showed no uranium. The initial concentrate was reduced to as low a volume as practical without crystallization and then treated as described above for the high uranium content solutions.

Throughout the extraction process, care was required to avoid the formation of emulsion which would carry large amounts of impurity over to the uranyl nitrate. High initial acid concentration had to be avoided since the introduction of a large amount of $\text{Ca}(\text{OH})_2$ for pH adjustment contributed to emulsion formation. Furthermore, $\text{Cu}(\text{NO}_3)_2$ proved preferable to $\text{Al}(\text{NO}_3)_3$ for specific gravity adjustment in that it was less conducive to emulsion formation.

The procedure for cake formation and chlorination was the same as that used in plant-scale HCP process for formation of UCl_4 .

In plant-scale operations, final degassing of the UCl_4 was accomplished after it had been placed inside the charge bottle whereas with the quantity of material involved in these collections, the charge was degassed while contained in a fritted glass filter which had been sealed on the open end. The filter retained the UCl_4 yet allowed the occluded gas to escape. After completion of the degassing, the transfer of UCl_4 from the filter to the charge bottle was made in a dry box.

D. Salvage

Nothing was discarded from this recycle until it was determined that no uranium was present. Solutions containing nothing detrimental to

~~SECRET~~

~~SECRET~~

recycle operations were added to the dilute evaporator. Carbon parts from the units, residues, and paper salvage items were leached, ignited, and releached. Organic liquids and residues were extracted with dilute nitric acid. The liquid nitrogen trap was cleaned with chloroform from which the uranium was washed with nitric acid. This wash was added to the dilute solution. Detectable amounts of uranium were seldom present on the trap during either the exploratory or collection operations since the equipment was designed for maximum retention of uranium within the liner.

IV. Run Figures and Recovery Data

The first series of material balance runs was made without complete data on each phase of the operation. A measured amount of 700-cycle UCl_4 was charged to each run and figures were obtained on the recovery for each run. Eleven such runs were made and an average recovery figure of 84 per cent obtained.

A second series of three individual runs was made using 1300 recycle material and wash solutions from all runs were combined for recovery. Results on this series showed better than 100 per cent recovery.

A series of runs was also made using 1900 recycle material. As in the 1300 series, only one recovery was made. Recovery was 96.7 per cent.

At this point, analytical facilities were set up to cover every phase of operations for an additional series of four runs made with normal material. Samples for analysis were taken at the end of each step in the recycle in the hope that some light could be shed on the erratic results

~~SECRET~~

~~SECRET~~

which had been obtained. It was realized, however, that the one phase of the recycle about which practically nothing could be measured was still the largest potential source for loss. This was the unit washing process.

The results (Table 1) were fairly consistent through the four runs but still showed a large loss. The average recovery was 86.1 per cent.

Table 1

Average Material Balance for Four
Analytically Controlled Recycle Runs

(1) Input into cycle	11.6 gms U
(2) Uranium recovered	
A. Cake formation condensate	0.32 gms U
B. Cleanup of chlorination equipment	0.56
C. HNO ₃ extraction of chlorination organics	0.44
D. Condensate from dilute solution concentration	0.12
E. Raffinate from ether extraction	less than 1 ppm
F. Ether from extraction	0.26
G. Carbons	0.80
H. Uranyl nitrate solution	7.49
	<hr/>
	Total Recovered 9.99 gms U
	Per Cent Recovered 86.1

The average recovery for all nineteen material balance runs was 87.6 per cent; it is significant, however, that those runs in which per cent recovery was based on weights of uranium recovered gave consistently higher percentages than the runs which were based on analytical data. There were not enough runs based on weight to justify acceptance of the higher values. Table 2 gives the recovery averages for all material balance runs.

~~SECRET~~

Table 2Recovery Averages for All Material Balance Runs

<u>Series</u>	<u>Material</u>	<u>No. of Runs</u>	<u>Average Recovery Per Run</u>	<u>Type of Balance</u>
I	Normal	11	84.0%	Analytical
II	1300	3	100.0%	Weight
III	1900	1	96.7%	Weight
IV	Normal	4	<u>86.1%</u>	Analytical
Weighted avg. for 19 runs			87.6%	

The recovery figure quoted in the feasibility report was the actual average of the entire number of runs (Table 2). Accepting this value, it was estimated that six collections could be made. This is illustrated in Table 3.

Table 3Anticipated Loss Figures for a Six Collection Series

<u>Run No.</u>	<u>Starting Material (gms U)</u>	<u>Equipment Holdup (gms U)</u>	<u>Loss (gms U)</u>	<u>U For Charge</u>	<u>UCl₄ For Charge</u>
1	11.3	1.3	-	10.0	15.9
2		1.4	1.2	8.8	14.0
3		1.4	1.1	7.7	12.3
4		1.4	1.0	6.7	10.7
5		1.4	0.8	5.9	9.4
6		1.4	0.7	5.2	8.3

~~SECRET~~

These estimates were included in the feasibility report submitted to the Atomic Energy Commission and approval was granted to proceed with the actual collections.

No analytical work was done during the collection runs, with the exception of analysis of the solutions and residues which were to be discarded from the recycle. For this reason, no results are available on recovery during the collection runs except the final weight figure obtained after completion of the runs.

The recycle was started with 11.3 grams of uranium; and after eight collection runs had been made, 8.8 grams of uranium remained. This shows a total recovery figure, after eight runs, of 77.9 per cent. For comparison, extension of the method shown in Table 3 to eight runs indicates that 35.2 per cent would have been expected. The mass analyses of the material before and after the collections are presented in Table 4.

Table 4

Mass Analysis of 236-bearing Uranium³

	<u>Before Start of Recycle</u>	<u>After End of Recycle</u>
U-234	0.79%	0.81%
U-235	93.72	91.81
U-236	0.82	0.72
U-238	4.67	6.68

3. Y-12 Mass Analysis Laboratory, Roger Hibbs in charge.

~~SECRET~~

~~SECRET~~

APPENDIX H

~~SECRET~~

~~SECRET~~

APPENDIX H

Chemical Purification of U-236

Although the purification and analysis of uranium is a standard Y-12 procedure, experience with quantitative recovery and purification of milligram quantities of uranium was limited. As a result, a development program was initiated with known amounts of natural uranium to determine techniques, efficiency of recovery, and expected chemical purity of product.

As a result of this development program, the following innovations were found to yield satisfactory results:

1. Ammonium nitrate is superior to calcium or copper nitrate as a "salting out agent" for uranium. Ammonium nitrate is as quantitative as either of the two metal nitrates and has the added advantage of introducing fewer contaminants.

2. Of the two extracting agents tried, dibutoxytetraethylene glycol (penta ether) and diethyl ether, the first was the more quantitative.

3. Carbon tetrachloride was added to the "penta ether" from which mixture the uranium was easily extracted with water. This procedure replaced the commonly used ammonium sulfate washout of the ether layer, since it had been found that small amounts of uranium were lost in driving off sulfuric acid from ammonium sulfate. The necessity for maximum product recovery could tolerate no such losses.

Experience with natural uranium showed that with these changes from plant procedure, satisfactory results could be obtained; thus this method

~~SECRET~~

~~SECRET~~

was applied to U-236 samples without spectrochemical analyses due to the small quantity of material involved.

Upon receipt, the graphite uranium collector was broken into small pieces, placed in a 100 ml platinum dish, and ignited in the presence of excess oxygen at 850°C usually for 20 to 24 hours. After cooling, the dish was removed from the furnace and the sample put into solution with 10 ml of concentrated nitric acid and a few drops of 30 per cent hydrogen peroxide. The solution was then filtered since the samples always contained a small amount of insoluble material at this point. The filtrate was reserved and the filter paper was analyzed for uranium by fluorescence using a fluoride matrix. The uranium content was never found higher than two micrograms and this represented less than 0.25 per cent of the sample, hence no recovery was considered necessary.

The nitric acid filtrate was taken to dryness cautiously, and the dry salts redissolved in a saturated solution of ammonium nitrate containing 5 per cent nitric acid by volume. The resulting solution was transferred to a 60 ml separatory funnel along with 10 ml of dibutoxytetraethylene glycol ("penta ether") and shaken for at least 10 minutes. The two components were allowed to settle for several minutes and the aqueous layer drained away. Ten milliliters of carbon tetrachloride and 20 ml of distilled water were added to the ether layer and shaken for at least 10 minutes. After settling for at least an hour, the fractions were separated into 150 ml beakers. The ether layer was analyzed for uranium by fluorescence; and in no case, were more than two micrograms of uranium found.

~~SECRET~~

~~SECRET~~

The aqueous layer, now containing the uranium, was treated with concentrated nitric acid and hydrogen peroxide until all the organic material had been destroyed. The solution, approximately 5 ml, was transferred into a small tared platinum vessel for evaporation and ignition. The sample was ignited to constant weight at 850°C. The uranium oxide (U_3O_8) was then dissolved in a minimum of nitric acid and made up to a volume such that 1 ml would represent 100 micrograms of uranium. This 1 ml sample was transferred to the physical analysis group for assay and the remaining volume was reduced and pipetted into a glass tube which was then sealed, and shipped.

~~SECRET~~

~~SECRET~~

APPENDIX I

~~SECRET~~

APPENDIX I

Spectrographic Assay of Product Material

The problem of performing an assay on the small product level samples, enriched in U-236, was handled by means of a spectrographic assay made possible by the isotope shift observed in heavy elements.

I. Theory of Isotope Shift in Heavy Elements

The isotope displacement effect¹⁻⁶ in the spectra of the heavy elements is due principally to the penetration of the optical electron into the non-Coulomb force region at the nucleus. Thus, a strongly penetrating electron, such as the $s_{\frac{1}{2}}$ or the $p_{\frac{1}{2}}$ electron would be influenced more than a relatively non-penetrating electron, such as the $p_{\frac{3}{2}}$, d, etc. electron. The same type electron experiences different departures from the Coulomb inverse square law in the various isotope species. This isotope displacement effect is frequently referred to as a nuclear volume effect since the divergencies from the Coulomb law are basically due to macroscopic changes in the nuclear volume.

II. Spectro-Isotopic Assay of Samples Enriched in U-236

Isotopic displacements in the spectra of U-238 and U-235 were first reported in a document by Long and Smith,⁷ who pointed out its possible

-
1. Racah, G., Nature 129, 723 (1932).
 2. Rosenthal, J. E. and G. Breit, Physical Review 41, 459 (1932).
 3. Breit, G., Physical Review 42, 348 (1932).
 4. Kopfermann, H., Kernmomente, Leipzig (1940).
 5. Smorodinsky, J., Journal of Physics (USSR), 10, #5, 419 (1946).
 6. Schawlow, A. L., and M. F. Crawford, Paper K8, Meeting of the American Physical Society, April 28-30, 1949.
 7. Long, D. R. and D. D. Smith, "Observed Differences in the Arc Spectra of U-235 and U-238", B-4.380.121, February 12, 1947.

~~SECRET~~

~~SECRET~~

application to the assay of uranium materials. Further study of these isotopes of uranium and also of U-233, supported the correctness of qualitative mass assay by the method of emission spectroscopy.

The extension of the number of uranium isotopes studied by spectroscopic means has followed as a result of the research program underway in the Y-12 Isotope Physics Group. Preliminary data on the shifts in U-236 and U-234 presented in Figure 15 and Table 1, illustrate the isotope structure in the prominent 4244 Å spectrum line of uranium.

Table 1
Observed Isotope Shifts

<u>Isotope</u>	<u>λ</u>	<u>$\Delta \lambda$</u>	<u>$\Delta \sigma$</u>
U-238	4244.373 Å	0.000 Å	0.00 cm ⁻¹
U-236	4244.226	0.147 ± 0.010	0.82
U-235	4244.122	0.251 ± 0.002	1.39
U-234	4244.075	0.298 ± 0.003	1.65
U-233	4243.977	0.396 ± 0.005	2.20

The quantitative evaluation of the isotopic content of uranium materials presupposes sufficient isotope material to obtain a detectable spectrum line density. Sample sizes may vary from about 10 mg (in studying an isotope present at about 0.2 %) to as low as 20 micrograms (in studying isotopes present at greater than about 10%). Analyses of U-236 concentrates were carried out on approximately two micrograms of the U-236 isotope.

For analysis of U-236 enhanced material, approximately 20 micrograms of uranium in nitrate solution, as submitted by the Isotope Chemistry Group

~~SECRET~~

109

SCALE IN ANGSTROMS

FIGURE 15. ISOTOPE STRUCTURE OF URANIUM
LINE 4244 A

~~SECRET~~

in Y-12, were absorbed in an approximately $1/4 \text{ cm}^2$ filter paper, which was dried and charred under a sun lamp and then ashed in a platinum crucible at about 600°C . The ash of about 40 micrograms was transferred to the crater at the apex of a cone-shaped graphite electrode and sealed with collodion. The lower electrode (anode) was prepared by sharpening a $1/8$ inch diameter electrode in a pencil sharpener, flattening the cone apex, and drilling a $1/40$ inch diameter hole in the apex. The cathode was a sharpened $1/8$ inch graphite electrode. This is illustrated in Figure 16 which shows the electrodes, a pencil point, a 10 mg sample, and a sample of about 50 micrograms for comparison purposes.

The arc was operated in the conventional vertical position at 3.5 amperes d-c for about 15 seconds, after which time the contribution to the uranium spectrum was negligible. The electrode gap of about 6 mm was projected onto the step-sectored slit with approximately 1.5 magnification. These step-sectored sample pictures could not be used for intercomparison because of non-uniform illumination of the slit. However, the steps served to permit a choice of optimum line density conditions. A 21-foot Jarrell-Ash, 15,000 line per inch grating spectrograph was used in second order at a dispersion of about $4.8 \text{ \AA/mm}$.

The plates were calibrated by burning for 15 seconds a 20 milligram sample of 40 per cent U_3O_8 in graphite powder. Arc gap was 10 mm and the arc was placed about 8 feet from the slit giving, with the lens used, a magnification of about 15. The central portion of this magnified image was checked and found to give uniform illumination along the spectrum lines.

~~SECRET~~

FIGURE 16. SPECTROGRAPHIC ELECTRODES

~~SECRET~~

A 1.5 to 1.0 step sector was used.

The resulting spectrograms were densitometered and the relative intensities of the three isotope components determined by straightforward spectrographic procedures (U-234 was assumed zero). The percentage concentrations (corrected for background and overlap of components) were then determined by taking ratios of the isotope component intensity to the total intensity of the three components. Table 2 summarizes the results of this analysis:

Table 2
Results on Assay of Samples Containing U-236

<u>Sample</u>	<u>U-235</u>	<u>U-236</u>	<u>U-238</u>
P-26-R3	61%	18%	21%
P-26-R4	54%	29%	17%
P-26-R5	68%	21%	11%
P-26-R6	58%	20%	22%

The accuracy of these determinations is approximately $\pm 10\%$ of the value listed for both U-235 and U-238, but only $\pm 20\%$ for the U-236 because of overlap with the U-235. Figure 17 illustrates one of the exposures to the U-236 material compared with a previously obtained series of isotopic pictures.

The major limitation on the accuracy of these determinations is the imperfect separation of the 235 and the 236 components. If larger samples could have been made available, the 236 determination could have been considerably improved, and also the determinations of both 235 and 238.

~~SECRET~~

$$\text{U}^{238} - 4244.37 \text{ \AA}$$

$$20\% \text{ U}^{235}$$

$$40\% \text{ U}^{235}$$

$$60\% \text{ U}^{235}$$

$$80\% \text{ U}^{235}$$

$$95\% \text{ U}^{235}$$

$$\text{U}^{233}$$

$$\frac{1}{3} \text{U}^{233} \quad \frac{1}{3} \text{U}^{235} \quad \frac{1}{3} \text{U}^{238}$$

$$\frac{7}{10} \text{U}^{235} \quad \frac{2}{10} \text{U}^{236} \quad \frac{1}{10} \text{U}^{238}$$

FIGURE 17. U-236 SPECTROGRAM

~~SECRET~~

A 500 microgram sample would have been sufficient to permit the use of a large 21 foot, 20,000 line per inch grating spectrograph at a resolving power exceeding 100,000 in second order.

~~SECRET~~

APPENDIX J

APPENDIX J

Health Hazards

Aside from the toxic effect of uranium, which characteristically manifests itself by degenerative changes in the kidney, the real point to be considered in the safe handling of material in which U-236 is concentrated is its relatively high alpha activity.

The specific activity of pure U-236, i.e., the number of disintegrations per gram per second, is somewhere between that of U-234 and U-235 (2.12×10^8 and 8.02×10^4 d/gram/sec., respectively). This amount of activity obtained from enriched uranium, especially with respect to U-234 and U-236, prescribes the need for a different concept of the hazards involved. Unlike the safeguards involved in the handling of normal uranium, where it can be treated much as any other chemical poison, this material decrees safe handling in full view of the acute alpha hazard.

The heavy alpha particle penetrates only relatively short distances before all of its energy is expended in producing low energy secondary ions. These, in turn, produce more ions until all the energy is converted into energy of chemical dissociation and heat. The fact that it is a densely ionizing particle simplifies the principle of reducing the hazard to a safe level.

Shielding against alpha particles presents no particular problem if the emitter is a solid or liquid. The range of an alpha particle, or the distance traveled before being stopped, depends on the number of collisions per unit

~~SECRET~~

path length. In air, a 5.5 Mev alpha travels about 4 cm; the same particle would travel only about 0.002 cm in aluminum. A sheet of paper, or the horny layer of the skin stops them completely. In general, the depth of penetration of alpha particles on any solid material is extremely small.

The biological effect of alpha radiation on the external surfaces of the body, therefore, is entirely negligible. Internally, however, the densely ionizing alphas are most effective in producing damage because all of the ionization occurs in thin layers of sensitive tissues. A simple calculation shows that only about 1.2×10^7 alpha particles of 4 Mev per gram of tissue are required to produce a dose of one roentgen equivalent physical. Thus, in the handling of alpha active material, it becomes necessary to design equipment and prescribe safety precautions with a view to preventing its entry into the body.

If alpha emitters get into the body, they will concentrate at places determined by the chemical nature of the material and will produce injury at those sites. Uranium, like radium and plutonium, is a bone-seeking alpha emitter. Once within the body, uranium tends to complex with the phosphates of the bone in a thin layer between the marrow and the shaft of the bone and produces damage almost entirely by the action of the emitted alpha particles. This complexing reaction takes place very rapidly following exposure, and the deposited uranium is excreted very slowly from the body.

Due to the nature of operations in separating uranium isotopes by the electromagnetic method, the chief danger appears to be that of airborne uranium. An alpha-emitting, airborne, finely divided dust is a particularly

~~SECRET~~

~~SECRET~~

dangerous material because of the ease with which it can be absorbed by inhalation. In handling such materials, the safety precautions must be adequate to take care of the worst possible accident, not merely sufficient to cope with a routine operation. An adequate ventilation system is essential, but one cannot overlook the fact that ventilating systems, like all mechanical devices, are subject to failure and that supplementary precautions may be necessary. These may take the form of regularly worn respirators and easily accessible gas masks with a self-contained oxygen supply. Enough instruments should be available to insure a continuous and complete monitoring of the hazard, so that any accident may be recognized as soon as it occurs. Moreover, scrupulous care must be taken to avoid spilling any of the material and to avoid entrance into the body through cuts or through careless manipulation as may occur in pipetting.

Standard precautions in the handling of alpha-emitting materials include the following:

1. All pipetting must be done by mechanical aids.
2. Rubber gloves must be used whenever hand contamination is possible, with regular surgical techniques used for application and removal.
3. Adequate respirators must be worn whenever alpha emitters may be present in room air in concentrations greater than $10^{-8} \mu\text{c}$ per liter.
4. Food must not be permitted in areas where active materials are used.
5. Hands must be thoroughly washed and monitored before eating, smoking, or leaving work.

~~SECRET~~

~~SECRET~~

In determining the degree of exposure, and perhaps the amount of uranium stored in the body, various tests of the urine and feces are made. These tests are not easy because calculations must be made from measurements of the radioactivity found in these body wastes. In the case of uranium, for the period following the first 72 hours after exposure, the rate of excretion is very low, and measurements must be made on a very low activity basis. Moreover, a further complication arises from the fact that the ratio of retained to excreted uranium varies with the time following the intake. All of which means that entire 24 hour voidings must be collected and elaborate precautions in sampling technique must be followed to permit a study of the effect of these unknown factors.

During the course of this project, the personnel involved and their work stations were carefully observed by the Health Physics Group using standard monitoring techniques. Fortunately, all handling of the active material was restricted to the four chemists who performed the recovery operations and the attendant analytical work. Their cooperation is largely responsible for the success attained in reducing potentially hazardous operations to safe working levels.

All work areas were restricted to personnel directly involved in the project. These areas were meticulously "policed" on the assumption that contamination was inevitable; hence, frequent and routine cleanings were carried out. To reduce the likelihood of cross contamination by tactile transmission still further, shoe covers were added to the complement of protective clothing usually prescribed in such cases, i.e., uniforms, caps,

~~SECRET~~

~~SECRET~~

gloves, and respiratory protective devices. Proper disposition of these items during the work day together with daily clothing changes insured excellent control of the potential dangers which might result from contamination of the areas and the personnel.

The different steps in the process are briefly discussed with a general statement regarding the possibilities of exposure:

Preparing the charge: Charge preparation was essentially a wet chemistry and closed cycle process. With adequate hood facilities available there were no exposures demonstrated.

Removing calutron from tank following run. This operation was accomplished within a period of two to three minutes and, while measurable quantities of airborne uranium were detected, the likelihood of exposure was judged to be slight.

Disassembly and washing of calutron units. These operations were performed in an area where local ventilation facilities were admittedly inadequate or nonexistent. The exposures demonstrated here proved to be the most dangerous. The analyses of respirator filters used at this site would usually show activity of the order of several hundred counts per minute. Apparently the combination of mechanical scrubbing along with the use of hot water caused the surrounding air to become contaminated by entrainment.

Recovery of product material from receiver. All of the recovery operations, i.e., scraping, burning, extracting, etc., were performed in an adequately equipped laboratory. Careful techniques were employed throughout the course of these recovery operations and possibilities of exposure to

~~SECRET~~

~~SECRET~~

personnel proved to be very slight.

The chemists involved in this project subjected themselves to a routine program of daily urine analyses at the start. Excretion findings soon indicated that checks to this extent were unnecessary. It was determined that the analyses of 24 hour voidings were necessary only on days when active material and contaminated equipment were handled. The analytical results were spotty and indeterminate. There seemed to be just fair correlation between urine concentrations and suspected time of intake. It is known that the analytical procedure for determining uranium in urine is relatively insensitive to very low concentrations of the active material and the indeterminate results can undoubtedly be attributed to this inadequacy. Certainly it can be said that demonstrable absorption of uranium during the course of this project was in the range of lower sensitivity limits obtained in present methods and techniques of exposure monitoring. These limits being considerably below the accepted tolerance levels, it is therefore safe to conclude that no over-exposures to personnel were obtained during the project.

~~SECRET~~

~~SECRET~~

APPENDIX K

~~SECRET~~

APPENDIX K

Final Runs and Operations

The last exploratory run was terminated October 27, 1948, and the unit was washed in preparation for collecting U-236. When required for thorough cleansing, parts were rewashed to minimize dilution of the U-236 bearing material by natural uranium. Upon completion of the wash, the unit was reassembled, worn parts being replaced where necessary, and prerun checks such as water flow and vacuum tests were conducted in the tank.

Upon completion of assembly and preliminary testing, three short runs were made using nitrogen only to determine insofar as possible whether electrode alignment was satisfactory. The first of these runs failed to yield satisfactory beam resolution due to electrode misalignment. This condition was corrected with the result that successive test runs were normal.

I. Collections

On November 15, 1948, the unit was visually checked; and the production charge, consisting of 16.5 grams of UCl_4 , was installed. The following startup and operating procedure was followed:

1. Magnetic field current was set at 515 amperes giving approximately 6,200 oersteds field strength.
2. A rate of rise of 0.0005 microns per second or less was obtained.
3. Maximum allowable tank base pressure was 0.015 microns with no arc support gas being admitted.
4. The filament was set at 3.9 amperes primary.

- ~~SECRET~~
5. Accelerating voltage was set to approximately 9.5 kv and arc voltage applied. Pressure limit 0.15 microns.
 6. An arc was struck with nitrogen.
 7. Arc casting heaters were turned on (1.2 amperes primary maximum), and temperature controller was set at 250°C.
 8. After pressure dropped below 0.02 microns, the oven heater was turned on (0.5 amperes primary maximum) and the micromax set at 300°C.
 9. Temperature was increased as required to obtain best operation.
 10. The run was to be terminated if no satisfactory results had been obtained when the temperature reached 500°C.

In case of an internal water leak the following procedure was to be followed:

1. Turn off water.
2. Disconnect source and liner cooling water lines.
3. Let tank down to air.

If the unit was to be shut down for more than 15 minutes, the temperature was to be set back to 300°C; and after charge installation, the unit could be removed only if a chemist was present.

The expected total ion current was 450 - 700 microamperes, and a ratio between the peak U-235 to U-236 ion beams of 35 to 1 or greater was required before the shutter could be opened. The ratio after the shutter was opened was required to be 12 to 1 or greater with steady operation.

~~SECRET~~

~~SECRET~~

The first run was unsuccessful in that steady operation was not obtained nor was the U-235 to U-236 beam ratio acceptable. In addition, sidebands were strong and source drains were unusually high. Post run inspection revealed that the unit was clean and the electrodes were in good alignment, but the charge bottle vapor exit was blocked with a hard, coarse crystalline deposit. The material remaining in the charge bottle proper appeared normal; and since only six grams had been used, no charge recycle was required. Reasons appearing to contribute to failure included (1) excess charge in the bottle, (2) too rapid heating, and (3) insufficient heating in the arc and vapor manifold regions. All probably contributed; but subsequent findings indicated that insufficient arc casting heating was the largest single factor.

The second production run was attempted using the ten grams of charge remaining from the first run plus two grams of new material. Operating procedure was varied in that extreme care was exercised in temperature control to avoid overheating. Again, low outputs and inadequate beam ratios were obtained, and arc casting heater failure due to overloading led to termination of the run. Inspection revealed that the vapor manifold was again plugged with condensed charge material indicating insufficient heat in the arc casting region. All other source parts appeared normal.

At this point, an exhaustive check of heater power supplies was made. Both oven and arc casting heater supplies were loaded with actual heaters, and power output ratings were determined for each transformer tap setting. Upon completion of these tests, checks were made in which the source was

~~SECRET~~

~~SECRET~~

equipped with thermocouples in the vapor manifold and charge bottle to determine heater power-temperature relationships.

The results of these experiments indicated that heat "feed back" to the charge bottle was no problem, and that 500 watts power had to be supplied to the arc casting heaters to maintain vapor manifold temperatures in excess of 500°C. On the basis of these findings, it was decided to furnish unregulated power to the arc casting heater from an available manually controlled heater supply.

The heat transfer experiments, in addition to determining the power load for the source, revealed the importance of a small change in the arc casting fabrication. To facilitate manufacture, the mounting tabs were cast on the arc casting instead of being hard soldered with phos-copper as formerly. This change increased the radiating surface by only 5 per cent; but the transfer by conduction was markedly increased by elimination of the hard solder joint which had served effectively as a heat shield. This increased heat loss lowered the vapor manifold temperature below the charge condensation point; whereas, with castings having soldered-on lugs, acceptable temperatures had been just attainable with existing heaters and power supplies. Therefore, the mounting tabs were cut from the arc casting and reattached with phos-copper to reduce the conduction heat loss.

Incorporating the changes in equipment and operational techniques indicated from the heat transfer experiments, Run 3 was begun November 30, 1948, using a charge of 14.1 grams UCl_4 . This run yielded 9 hours innage with an average monitored ion current of 29.48 microamperes in the U-236 pocket.

~~SECRET~~

~~SECRET~~

The average ion currents monitored on the 235 and 234 probes were 500 and 167.3 microamperes, respectively. After innage was attained, operations were smooth with few adjustments being necessary; termination was caused by charge exhaustion.

From monitored ion currents, estimates of product quantity and isotopic composition were:

Estimated collection in the U-236 pocket: 2.36 mg

Estimated isotopic composition:	U-238	22%
	U-236	23.8%
	U-235	54.0%
	U-234	0.2%

The fourth production run was started December 10, 1948. Innage was obtained at 395°C, and the temperature was raised 2° per hour until just before termination. Arc, tank, and temperature adjustments were extremely critical prior to the attainment of innage, but operation was smooth after proper temperature was reached. Twelve hours of innage was obtained from this run; and average ion currents in the 236, 235 and 234 mass positions were 25.1, 550, and 120 microamperes, respectively. Termination resulted from charge exhaustion.

Based on monitored ion currents, the following estimates were prepared:

Estimated collection in the U-236 pocket: 2.67 mg

Estimated isotopic composition:	U-238	18.1%
	U-236	30.6%
	U-235	51.1%
	U-234	0.2%

~~SECRET~~

~~SECRET~~

The fifth collection run began December 15, and was very similar to Run 4 operationally. Innage time obtained was 7-3/4 hours, and average ion current at the 236 mass position was 25.88 microamperes. Average ion current at the 235 mass position was 550 microamperes; and at the 234 position, 124.6 microamperes. Termination was for receiver shorts. The estimated total uranium collected in the U-236 pocket was 1.78 milligrams containing:

U-238	18.3%
U-236	29.7%
U-235	51.8%
U-234	0.2%

The sixth production run was similar to the fifth. Average ion currents for U-236, U-235, and U-234 were 25.3, 450, and 94 microamperes, respectively. This run also terminated for receiver shorts after 11-1/2 hours of innage. An estimated 2.58 mg of uranium was collected in the U-236 pocket, containing:

U-238	14.0%
U-236	24.8%
U-235	60.9%
U-234	0.3%

All collections made up to this time used a U-236 entry slot 0.025" inches in width; however, in effort to increase the isotopic purity of the product, slot width for the seventh run was reduced to 0.015 inches. Innage time for Run 7 was quite short - 5-5/8 hours - due to a small charge; termination was due to charge exhaustion. An estimated 0.35 mg of uranium was

~~SECRET~~

collected with the following isotopic composition:

U-238	24.1%
U-236	33.7%
U-235	42.0%
U-234	0.2%

Run 8 began January 24, 1949, using the same type receiver as Run 7. Again innage time was short (5-3/4 hours) due to early charge exhaustion. Observed ion currents for U-236, U-235, and U-234 were 13.4, 400, and 40.0 microamperes, respectively, and were the basis for making the following estimates:

Amount of uranium collected: 0.68 mg

Estimated isotopic composition:	U-238	6.7%
	U-236	26.1%
	U-235	66.9%
	U-234	0.3%

No spectrographic assay was made on material collected in either Run 7 or Run 8 due to the small amount collected. No further collections were attempted since the AEC request had been exceeded with respect to both quantity and quality. Table 1 summarizes information on all collections including estimated and spectrographic isotopic compositions, average ion currents, and reason for termination.

II. Methods of Estimating Product Purity

Methods of calculating product isotopic compositions from observed ion currents were based largely on the bismuth beam shape obtained early in the

Table 1

Summary of Collection Run Data

Run Number	2601	2602	2603	2604	2605	2606	2607	2608
Date Started	11-15-48	11-18-48	11-30-48	12-10-48	12-15-48	12-29-48	1-11-49	1-24-49
Innager Time (Hours)	0	0	9	12	7-3/4	11-1/2	5-5/8	5-3/4
Reason For Termination	No output	J heater failure	Charge Exhaustion	Charge Exhaustion	Receiver Shorts	Receiver Shorts	Charge Exhaustion	Charge Exhaustion
Average Current (236 Position)			29.48 μ a	25.10 μ a	25.88 μ a	25.30 μ a	7.00 μ a	13.40 μ a
Average Current (235 Position)			500.0 μ a	550.0 μ a	550.0 μ a	450.0 μ a	270.0 μ a	400.0 μ a
Average Current (234 Position)			167.3 μ a	120.0 μ a	124.6 μ a	94.0 μ a	71.0 μ a	40.0 μ a
Estimated Quantity Collected			2.36 mg	2.67 mg	1.78 mg	2.58 mg	0.35 mg	0.68 mg
Estimated Composition of Collected Material (monitored)	U-238 U-236 U-235 U-234		22.0% 23.8% 54.0% 0.2%	18.1% 30.6% 51.1% 0.2%	18.3% 29.7% 51.8% 0.2%	14.0% 24.8% 60.9% 0.3%	24.1% 33.7% 42.0% 0.2%	6.7% 26.1% 66.9% 0.3%
Actual Quantity Collected (Recovered)			2.2 mg	3.2 mg	2.5 mg	2.8 mg	0.8 mg	0.9 mg
Revised Estimate of Composition of Collected Material	U-238 U-236 U-235 U-234		21.7% 23.2% 54.7% 0.2%	13.9% 26.3% 59.5% 0.3%	13.7% 20.0% 66.0% 0.3%	13.0% 22.2% 64.5% 0.3%	13.2% 15.2% 71.1% 0.5%	6.2% 19.9% 73.5% 0.4%
Spectrographic Assay of Collected Material	U-238 U-236 U-235		21.0% 18.0% 61.0%	17.0% 29.0% 54.0%	11.0% 21.0% 68.0%	22.0% 20.0% 58.0%	No Assay	No Assay
Enhancement	U-236		26.7	49.8	32.2	30.6	21.7	30.0
Receiver Slot Width	.025"	.025"	.025"	.025"	.025"	.025"	.015"	.015"

~~SECRET~~

12-inch radius calutron development program as well as on still earlier studies. The method used for calculating the amount of material deposited in the U-236 collector, which is based on electrochemical equivalents, is straightforward and has been used widely in the Y-12 Plant.

Calculation of a product isotopic composition estimate, however, presented a more unique problem. This method was based on the assumption that the contributions of other "beam tails" to the large observed U-235 ion current were negligible. On this assumption and on observed ion currents, the contributions of other beams to the small observed U-236 ion current were calculated from ratios of the isotopic abundances in the starting material with respect to U-235. This step yielded a calculated U-236 ion current which was, in turn, the sum of all contributing ion beams, and from which an isotopic estimate was easily derived.

For sake of illustration, complete calculations are presented for the fourth collection run. During this run, the average ion current in the 236 mass position and total innage time, taken from the recording meter chart, were 25.1 microamperes and 12 hours, respectively. Then,

25.1 microamperes x 12 hours = 301.2 microampere hours ion current,
and
microampere hours x 8.88 $\mu\text{gm}/\mu\text{amp hr.}$ = 2.68 mg U collected.

The average ion currents in the 234, 235, and 236 mass positions were:

234 position = 120.0 microamperes

235 position = 550.0 microamperes

236 position = 25.1 microamperes

~~SECRET~~

~~SECRET~~

The starting charge material had the following isotopic composition:

U-234	0.79%
U-235	93.80%
U-236	0.82%
U-238	4.62%

Abundance ratios for each isotope with respect to U-235 were:

U-234/U-235	- 1/118.73
U-236/U-235	- 1/114.39
U-238/U-235	- 1/20.30

Since U-235 was so predominant (93.80%), the assumption was made that "tails" of other ion beams falling on the probe at the 235 mass position were negligible. However, the contribution of U-235⁺ to the observed ion current in the 234 mass position was the recorded current (120 microamperes) minus the ratio of the U-235⁺ ion current (550 microamperes) divided by ratio of the starting abundance of the two isotopes (118.73):

$$120 - \frac{550}{118.73} = 120 - 4.63 = 115.37 \text{ microamperes of U-235}^+ \text{ monitored on the 234 position probe.}$$

This step was justified on the basis of the early work with bismuth in the development of the 12-inch radius calutron and still earlier beam studies. These studies had shown that beam shapes were similar in that the width of focus was the same at the 180° position regardless of beam intensity, and that beam intensity was a function of natural abundance. Thus, the measured beam currents were directly proportional to relative starting abundance of the isotopes. Dividing the peak U-235 ion current by the calculated U-235⁺

~~SECRET~~

~~SECRET~~

component at the 234 mass position gave an estimate of the intensity of a beam 1 mass unit away on the low energy side; i.e.,

$$\frac{550}{115.37} = 4.77$$

Assuming this ratio to be doubled two mass units away on the low energy side, the U-238⁺ component of the observed U-236 ion current was obtained:

$$550/20.30 = 27.09 \text{ microamperes Peak U-238 ion beam}$$

$$27.09/(2 \times 4.77) = 2.84 \text{ microamperes U-238}^+ \text{ in the U-236 pocket.}$$

In like manner, the U-236⁺ portion of the current observed at the 236 mass position was calculated:

$$550/114.39 = 4.81 \text{ microamperes U-236}^+ \text{ in the U-236 pocket.}$$

These calculated components of the observed U-236 ion current required correction, however, since each was derived from the observed U-235⁺ ion beam which was monitored through a 0.015" x 3" receiver defining slot. The ion entry slot at the 236 mass position was 0.025" x 3"; thus the ratio of the slot areas is 1.6 to 1 so the calculated U-238 and U-236 currents must be multiplied by the correction factor of 1.6.

$$2.84 \times 1.6 = 4.54 \text{ microamperes U-238}^+ \text{ in the U-236 pocket}$$

$$4.81 \times 1.6 = 7.70 \text{ microamperes U-236}^+ \text{ in the U-236 pocket}$$

It was assumed that the contribution of U-234⁺ to the total beam in the 236 position was so small that its omission would be compensated for by errors made in averaging ion currents from recording meters and run data. Thus, adding the corrected calculated values for U-238⁺ and U-236⁺ at the 236 mass position and subtracting their sum from the metered value, the contribution of

~~SECRET~~

~~SECRET~~

U-235⁺ to the total observed at the 236 position was obtained, i.e.,

$$25.1 - (4.54 + 7.70) = 25.1 - 12.24 = 12.86 \text{ microamperes U-235}^+$$

in U-236 pocket.

The ratio between the peak U-235⁺ and the calculated U-235⁺ at the 236 position was:

$$550/12.86 = 42.77$$

Doubling this ratio as before for two mass unit separation and applying it to the U-234⁺ calculated ion current, the U-234⁺ component of the metered current at the 236 position was determined:

$$4.63/(2 \times 42.77) = .05 \text{ microamperes.}$$

Adding the calculated components of the observed current gives:

U-238 ⁺	=	4.54 microamperes
U-236 ⁺	=	7.70 microamperes
U-235 ⁺	=	12.86 microamperes
U-234 ⁺	=	0.05 microamperes
Total		<u>25.15</u> microamperes

The percentage of the total ion current of each ion beam was calculated from these data yielding the following calculated isotopic composition:

U-238	18.05%
U-236	30.62%
U-235	51.13%
U-234	0.20%
Total	<u>100.00%</u>

~~SECRET~~

~~SECRET~~

The collected material from the fourth run was weighed after chemical recovery and found to be 3.2 mg instead of 2.67 mg as estimated from calutron operations. The estimate of product purity was revised upon receipt of this information and was based on the assumption that the extra material was of the same isotopic composition as the charge material.

$$3.2 \text{ mg} - 2.67 \text{ mg} = 0.53 \text{ mg U}$$

This extra material contained:

$$0.53 \times 0.0462 = 0.0245 \text{ mg U-238}$$

$$0.53 \times 0.0082 = 0.0043 \text{ mg U-236}$$

$$0.53 \times 0.938 = 0.4971 \text{ mg U-235}$$

$$0.53 \times 0.0079 = 0.0042 \text{ mg U-234}$$

These quantities were added to the originally estimated quantities of each isotope as follows:

$$\text{U-238} = 4.54 \times 12 \times 8.88 = 54.48 \times 8.88 = 483.78 + 24.5 = 436.28$$

$$\text{U-236} = 7.70 \times 12 \times 8.88 = 92.40 \times 8.88 = 820.51 + 4.3 = 824.81$$

$$\text{U-235} = 12.86 \times 12 \times 8.88 = 154.32 \times 8.88 = 1370.36 + 497.1 = 1867.46$$

$$\text{U-234} = 0.05 \times 12 \times 8.88 = 0.6 \times 8.88 = 5.53 + 4.2 = 9.73$$

Micrograms Total	<u>3138.28</u>
------------------	----------------

The revised estimate for isotopic composition was:

$$\text{U-238} \quad 436.28/3138.28 \times 100 = 13.9\%$$

$$\text{U-236} \quad 824.81/3138.28 \times 100 = 26.3\%$$

$$\text{U-235} \quad 1867.46/3138.28 \times 100 = 59.5\%$$

$$\text{U-234} \quad 9.73/3138.28 \times 100 = 0.3\%$$

Total	<u>100.00%</u>
-------	----------------

~~SECRET~~

~~SECRET~~

The spectrographic assay of this particular sample showed the following composition:

U-238	17%
U-236	29%
U-235	54%
U-234	Not detected

A similar procedure for estimating product isotopic composition was followed for all other collections.

~~SECRET~~

~~SECRET~~

~~SECRET~~