

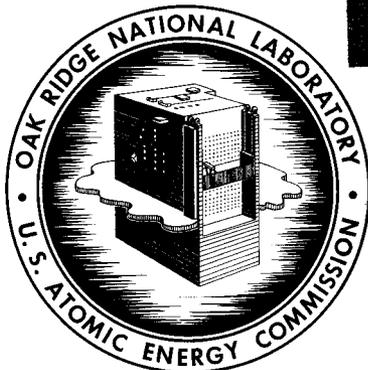


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TID-4500 (28th ed.)

REVIEW OF ORNL ELECTROMAGNETIC
SEPARATIONS PROGRAM
JANUARY - DECEMBER 1963

L. O. Love
W. A. Bell



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ISOTOPES DEVELOPMENT CENTER REPORT

REVIEW OF ORNL ELECTROMAGNETIC SEPARATIONS PROGRAM

JANUARY - DECEMBER 1963

L. O. Love, W. A. Bell

MAY 1964

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ABSTRACT

This report reviews developments and activities of the Electromagnetic Isotope Separations Department for 1963 in areas of magnetic fields, ion sources, ion receivers, charge evaluation, contamination, system modifications, quality control, and chemistry. Both stable isotope separations and special heavy element separations are included.

I. CALUTRON AND PROCESS DEVELOPMENT

The ORNL electromagnetic isotope separations program is the source of supply for >250 stable nuclides sufficiently enriched in isotopic content to be essential materials for nuclear research and technical applications. Limited quantities of enriched isotopes of Pu, U, and Th are also made available as Research Pool materials. Continued development efforts provide the improvements in equipment and techniques necessary to meet customers' specifications for isotopic purity and provide physical and chemical forms required for research applications.

In general the research field desires a constant increase in both isotopic purity and quantity of nuclides separated; however, some specific applications (i.e. dilution analyses) require very small quantities of ultra-pure isotopes. Other present and foreseen applications (i.e. construction materials for space programs) need quantities much larger than the separations program now provides but require an intermediate level of isotopic purity.

In separating isotopes by the electromagnetic method, ions formed in the source are accelerated over a short distance in the electrode region and then continue in free flight through a transverse magnetic field. The paths followed by these singly-charged ions through the magnetic field are semicircular, and the radius is defined by the relationship

$$ME = \frac{H^2 r^2}{(144.5)^2} ,$$

where

- M = mass of the ion,
- E = accelerating potential in volts,
- H = magnetic field strength in gauss,
- r = radius of curvature of the ion path in centimeters.

In theory, any separator having adequate control of E and H should produce a separation between two masses (e.g. M_1 and M_2) by dispersing the ion beams at the collector by a distance of $2(r_{M_1} - r_{M_2})$. Since the size of the calutron is fixed to accommodate a reference ion-path radius of 24 in., the dispersion produced between ions of heavy mass is inadequate to permit the use of optimized collector pockets. This limitation in collector design causes an undesirable loss of isotopic material through sputtering of previously deposited isotopes by incoming 35-kev ions. In addition to the sputtering loss, the mass resolution achieved is never 100% efficient; and isotopic purity requirements dictate the degree of precision which must be achieved in source performance, power regulation, vapor control, beam stabilization, and control of contamination.

Each calutron separation is approached with a dual objective: the accumulation of enriched isotopic samples for inventory and nuclear usage, and the acquisition of research and development data applicable to process improvement. An adequate understanding of some basic processes permits equipment and operating techniques to be conceived which will circumvent deleterious effects these processes have on the separation; in other instances, experimentation is directed toward acquisition of the knowledge needed for problem solution.

A. 255° Inhomogeneous Field Separator

Limitations imposed by inadequate beam dispersion in the 24-in. radius calutron and the desire to attain greater isotopic purity in a single-pass process were responsible for investigation of a separator designed to increase dispersion while retaining the advantageous features of the calutron. In theory, a separator achieving double-directional ion focusing through use of a circularly symmetric, radially inhomogeneous magnetic field which had an inhomogeneity factor of 0.5, a reference radius of 20 in., and a deflection angle of 255° would increase beam dispersion by a factor of 1.6 over that experienced in the 24-in. radius calutron.

Magnetic fields of this general type have been successfully used in mass spectrometry, beta spectrometry, and sector separators; and Russian scientists have developed a working 225° isotope separator using an inhomogeneous field. Published papers indicated that it should be possible to find a satisfactory inhomogeneous field for use in the ORNL separations program even though most of the investigations into the theory of inhomogeneous magnetic lenses had the objective of developing instruments other than isotope separators. To be effective in a separations program, an acceptable system would have the capacity to produce a well-focused image from an ion source having a comparatively long ion exit slit and large angular divergence of the ion beam. Past investigations had not completely covered these two aspects of the focusing problem.

Double-focusing separators require precise magnetic fields, and a computer program was established to determine a magnetic field which would minimize radial aberrations up to 3 in. away from the median plane. To a first order approximation this field is defined by the relationship

$$B_1 = B_0 \left(\frac{r_0}{r} \right)^n ,$$

where B_0 is the magnetic field strength at reference radius r_0 , and n is the inhomogeneity factor.

An inhomogeneous magnetic field is shown schematically in Fig. I-1. The field is circularly symmetric and the magnetic intensity decreases as the distance from the center increases. A field of high inhomogeneity decreases in intensity more rapidly than does a field of low inhomogeneity.

Compared with a 180° calutron system of unit dispersion, a field of 0.5 inhomogeneity has a focal angle of 255° and a dispersion of 2 whereas a field of 0.8 inhomogeneity has a corresponding focal angle of 400° and a dispersion of 5. Although the high dispersion value is desirable, focal angles >360° cannot be accommodated in isotope separations and reasonable compromises based on construction details must be accepted.

With inhomogeneous magnetic lenses, an ion traveling outside the magnetic midplane will be forced back toward the midplane by the action of the magnetic field. Thus an ion that begins its flight outside the midplane will oscillate through the midplane on its trip to the collector. A particle which leaves the source 2 in. from the midplane passes through this plane and strikes the collector at a point 2 in. from the midplane on the side opposite that from which it started. This point-to-point correspondence is referred to as double-focusing and provides a uniform ion current distribution at the collector of the 255° separator.

Evaluation of the magnetic field for the 255° separator was accomplished by programming the IBM 7090 computer to calculate the trajectory of any ion moving in a field of this type. This program made practical a high degree of freedom in choosing higher order field parameters and made it possible to incorporate certain refinements into the field shape which resulted in a more effective isotope separator. Results of this study showed that in addition to increasing mass dispersion, this type of lens also produced a more sharply focused beam than that achieved in the beta calutron. Under normal operating conditions, the focused beam in the 255° separator could be expected to have a width <0.040 in.

In adapting this type of system into a beta calutron, it was necessary to extend the useful magnetic field throughout as large a volume as possible because ion transmission would have to be limited to regions of accurately shaped magnetic field. By programming the computer to evaluate the potential distribution produced by an equipotential surface representing the magnetic pole pieces, it was possible to

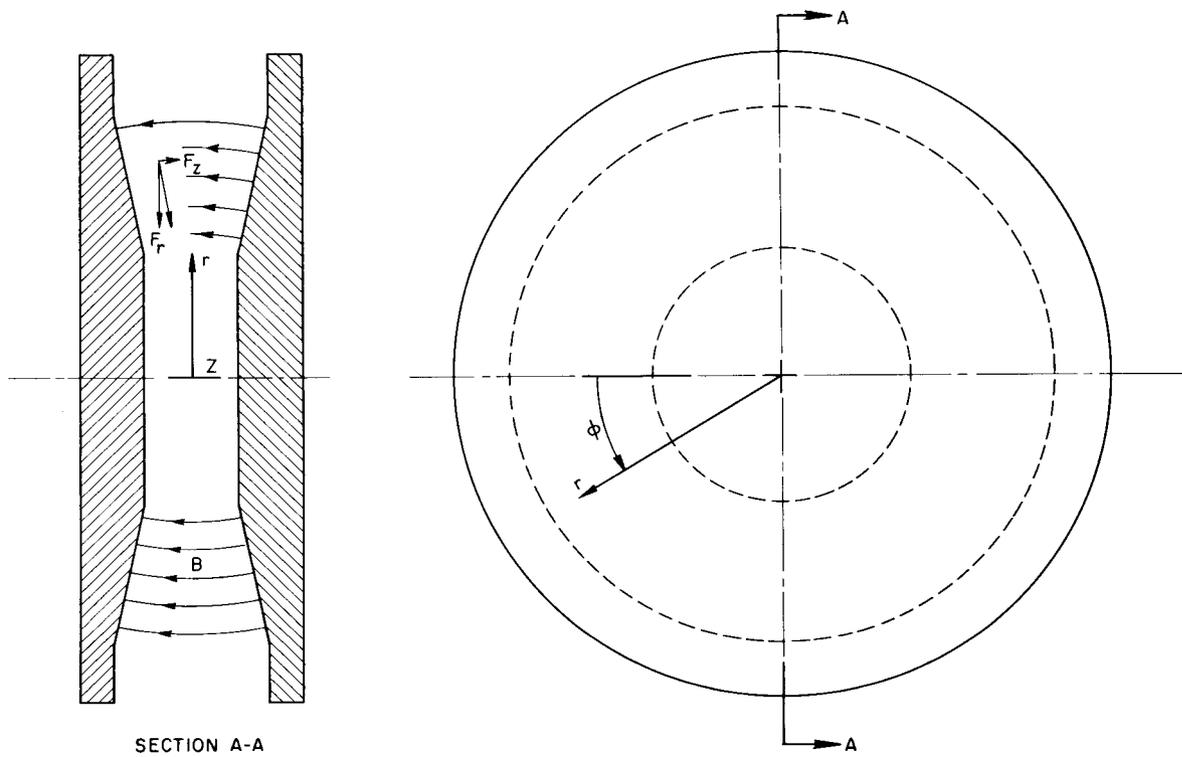
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Fig. I-1. Magnetic Field of the Type Used in the 255° Separator.

determine a contour which would produce a satisfactory magnetic field and still leave sufficient space in a calutron tank for a source, receiver, and liner. A cross section of the original magnetic shim determined by this technique is shown in Fig. I-2.

Incorporation of the magnetic shims in the calutron tank required changes in the source and collector. The new location of the source and collector in the tank is shown schematically in Fig. I-3. A source and collector modified to accommodate the 255° focal angle are shown in Figs. I-4 and I-5. Both the source and collector use many components which are also used in beta calutron equipment. The liner for a 255° separator, the only part of the system that was constructed entirely from non-calutron parts, was fabricated by forming copper sheets on a magnetic shim and fastening them to a stainless steel framework. The 255° system is completely interchangeable with the calutron concept and conversion from one system to the other can be completed in an 8-hr work day.

Although the 255° separator does nothing to alter one of the major causes of isotope contamination (beam instabilities), it does permit the separation of better isotopic samples because of its increased mass dispersion and improved imaging properties. Table I-1 compares results of separations completed in the 255° system with similar separations performed in calutrons and shows that the use of the 255° separator has resulted in significant improvement in isotopic purity achieved for the heavier elements.

B. Magnetic Fields

Sector Fields

Computer programming was established in conjunction with development of the 255° separator which made investigation of a variety of magnetic fields possible. Trajectories of ions in the calutron system were plotted to acquire a better understanding of focal problems which result when ions within the beam have abnormal initial velocity components, and studies were extended to include sector fields--both homogeneous and specific inhomogeneous types.

A sector-type separator could serve as a versatile unit satisfactory for beam studies, source evaluation, target preparation, and separation of high purity samples, particularly those second-pass operations using small quantities of pre-enriched materials. The high degree of versatility inherent in the sector instrument is due to the fact that both ion source and receiver are located outside the field of the analyzing magnet. Placement of these components in such a location dictates that ions enter and leave the main magnetic field through fringing fields, and evaluation of the action of these fields on the over-all ion trajectory is limited to approximation only.

In a uniform-field sector study the components of the fringing magnetic field can be calculated by means of a Schwarz-Christoffel transformation

Table I-1. Comparison of performance of calutron and 255° separator

Isotope	Purity achieved, %		Total current, ma	
	255° Separator	Calutron	255° Separator	Calutron
Cd ¹⁰⁸	88.6	84.0*	22	22
Cd ¹¹¹	98.4	97.8*	22	22
Cd ¹¹³	97.1	97.1*	22	22
La ¹³⁸	13.5	10.6*	7	9
¹ Hg ²⁰⁰	76.1	91.3*	4.1	2.8
¹ Hg ²⁰²	83.4	98.3*	4.1	2.8
Pb ²⁰⁴	86.6	81.0**	12	21
Pb ²⁰⁶	98.0	81.0**	12	21
Pb ²⁰⁸	99.1	96.0*	12	21
Ta ¹⁸⁰	0.47	0.30**	20	10
Sn ¹¹⁷	93.15	90.5**	22	22
Sn ¹¹⁹	93.16	91.7**	22	22

¹ 255° separator did not have special cooling on liner

* alpha (48-in. radius)

** beta (24-in. radius)

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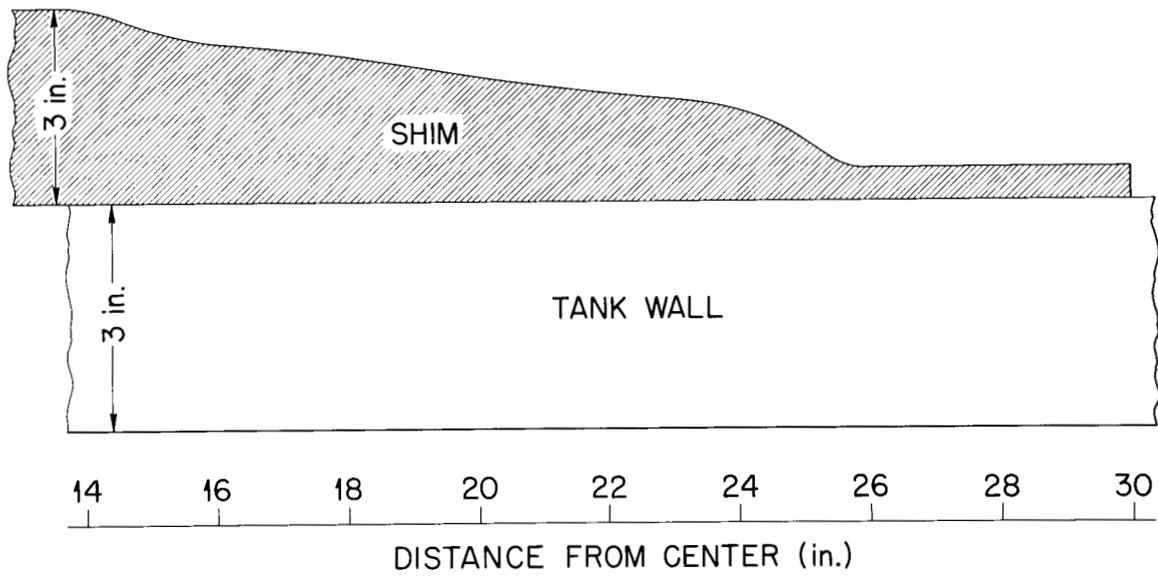


Fig. I-2. Contour of 255° Shim.

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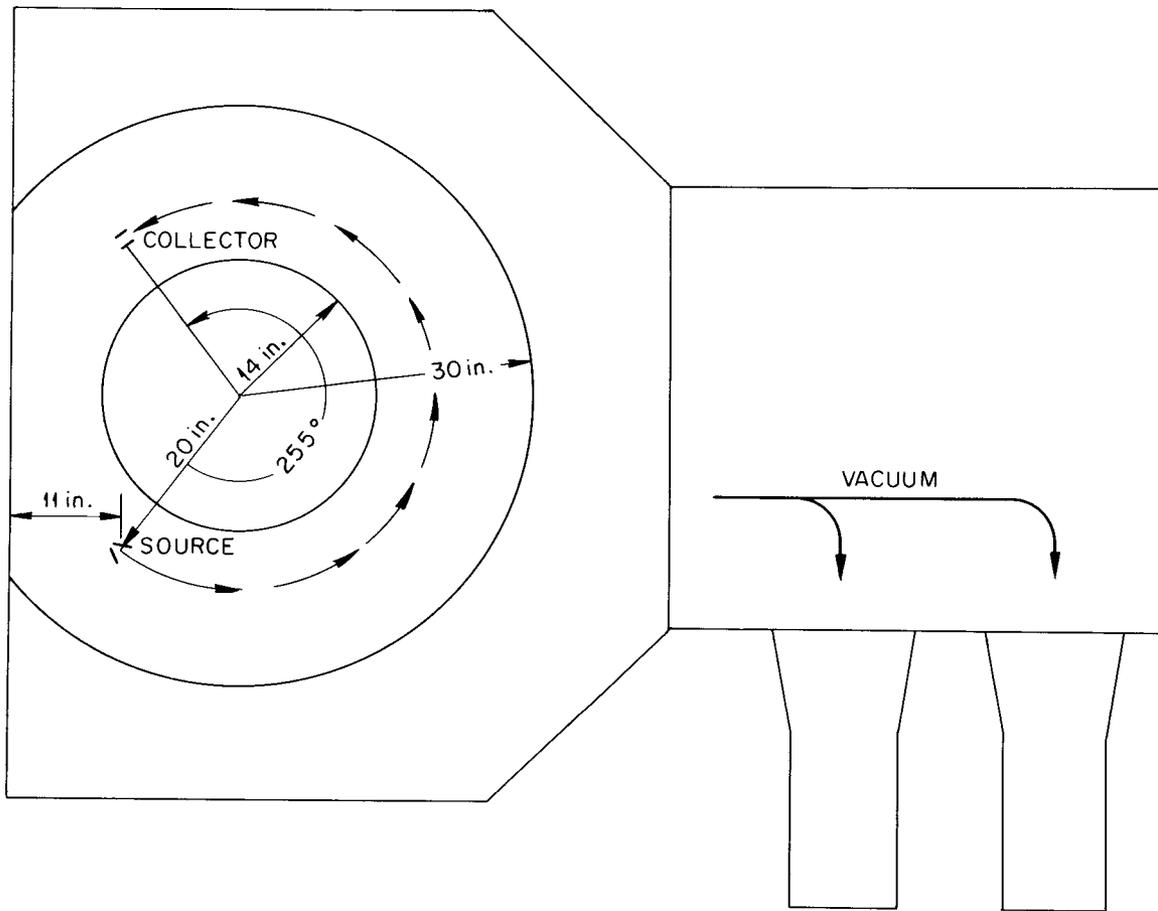


Fig. I-3. Shim Location in a Calutron Tank.

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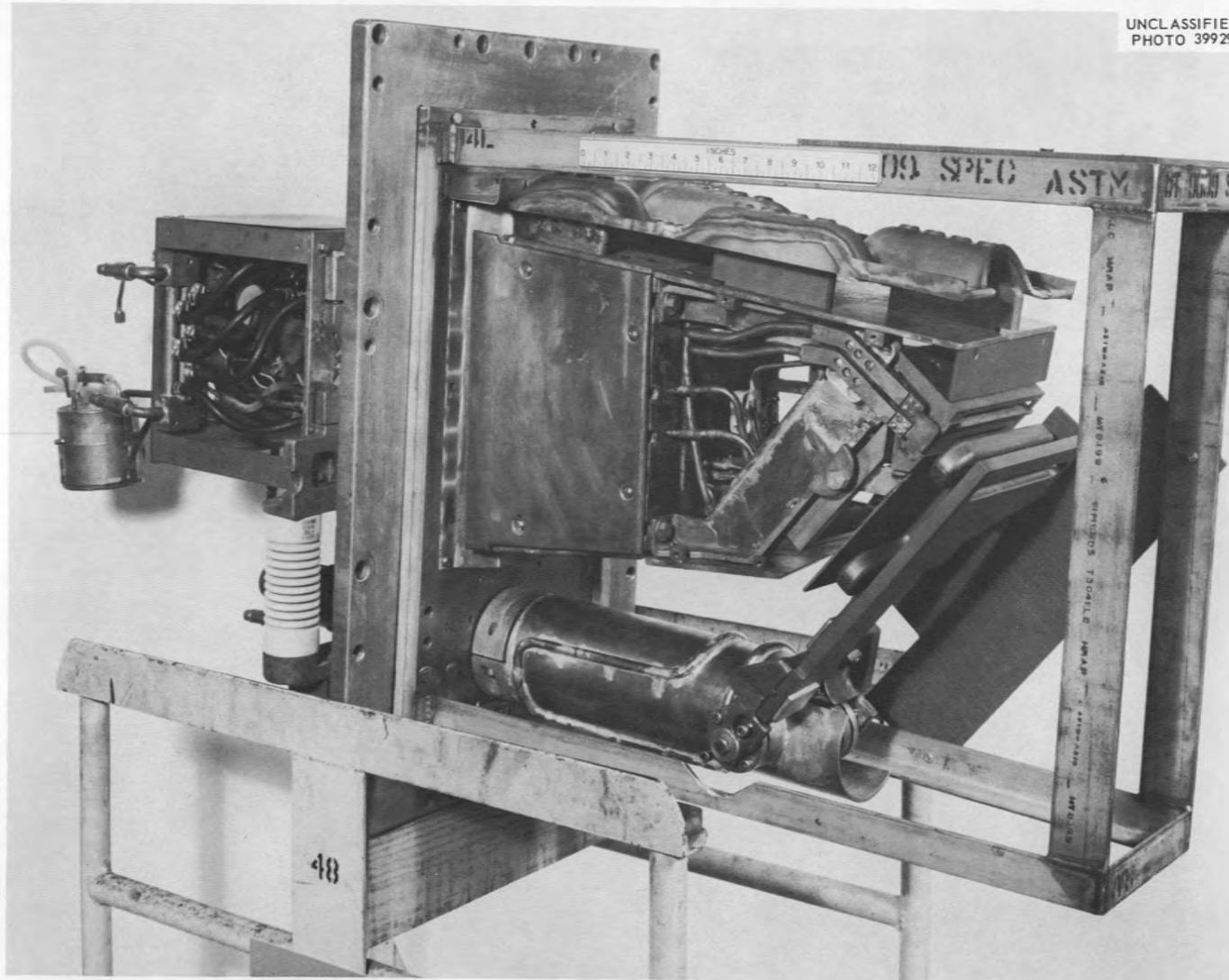


Fig. I-4. Ion Source Used in 255° System.

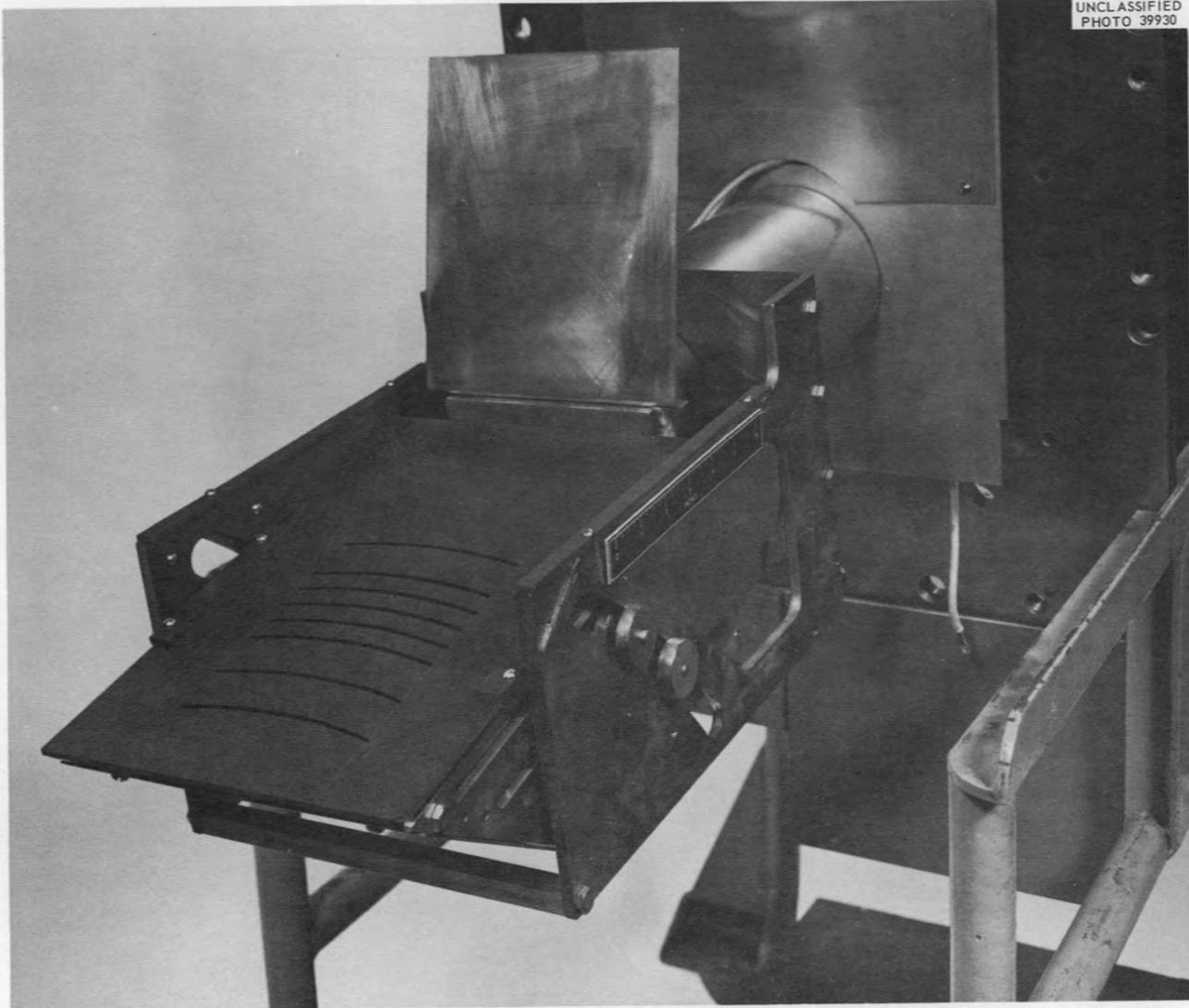


Fig. I-5. Receiver Used in 255° System.

in the complex z -plane. Unfortunately this technique cannot be applied directly to calculate the field of an inhomogeneous sector magnet. The approach that has been followed in the ORNL study of such fields is to estimate the effective field components by combining the field gradients from the fringing field in front of a uniform-field sector magnet with those of an inhomogeneous magnetic field with no fringing.

The 0.5 and 0.8 inhomogeneous field systems and the uniform field system have been investigated more extensively than other types or values of inhomogeneity, and either system can be utilized successfully in a separator. It is recognized, however, that control of initial ion conditions is more critical in inhomogeneous fields than it is in uniform fields, and the precision required in field formation increases with increasing inhomogeneity.

"Football" Shims

The focal pattern of ion beams in a conventional calutron is curved as a result of shimmed field action upon charged particles. The curvature necessitates the formation of correspondingly shaped collector pockets and defining plates; therefore, pocket construction is time consuming and costly.

In 1960 Balestrini and White¹ reported a system for use in shaping the magnetic field in a mass spectrometer which, if adaptable to calutron use, would produce a nearly straight focal pattern at the receiver. A set of shims of this type (called "football" shims because of their general shape) was fabricated using the generalized concept that a sharp cutoff in fringing field at half-gap width would be a reasonable assumption for calculating the needed shim profile. Shim installation (Fig. I-6) and subsequent field measurements revealed that this initial set of shims did not produce the desired field; however, a reference field was established from which refinements could be calculated.

In order to define the imaging properties of this type of lens, the field must be represented by an accurate expression in three dimensions. In a magnetic field with one plane of mirror symmetry, the normal component (B_z in this example) of magnetic flux density may be a function of x and y . If this quantity is known and is expressible mathematically, then it can be shown that the components in the surrounding volume are given by

$$B_z = \sum_{n=0}^{\infty} \frac{(-1)^n z^{2n}}{(2n)!} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right)^n B_z ,$$

¹S. J. Balestrini, F. A. White, Rev. Sci. Instr. 31(6): 633-6 (1960).

$$B_x = \sum_{n=0}^{\infty} \frac{(-1)^n z^{2n+1}}{(2n+1)!} \frac{\partial}{\partial x} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right)^n B_z, \text{ and}$$

$$B_y = \sum_{n=0}^{\infty} \frac{(-1)^n z^{2n+1}}{(2n+1)!} \frac{\partial}{\partial y} \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \right)^n B_z,$$

where B_z is the known expression for B_z as a function of x and y at the magnetic midplane. In theory, the preceding treatment would define the field components once B_z were known. In practice, the degree of convergence of the series could render the expressions useless in determination of the field components. In evaluation of the experimental calutron shims, it was necessary to use five terms to obtain sufficient accuracy for use in computer programming.

Examination of the measured field data (taken only at the midplane position) revealed that B_z could be expressed in the form

$$B_z = B_0 \left\{ 1 + \sum_{n=1}^3 A_n \exp \left[C_{xn} (x - x_n)^2 + C_{yn} (y - y_n)^2 \right] \right\}$$

when the constants were properly chosen. The three-dimensional trajectories of ions moving in this magnetic field were calculated from a computer program based on the above equations. The results of this work are shown in Fig. I-7 which gives the loci of points where scaled trajectories pierce an imaginary plane to form a focal pattern. This pattern is unacceptable for isotope separation, but improvements could result from further field shaping.

255° Inhomogeneous Field

The inhomogeneous field in the 255° separator provides double focusing of ions with point-to-point correlation between source and receiver. Original construction was based on the premise of one-arc operation and collection in slightly curved receiver pockets. Further study of ion trajectories in this field indicated that straightening of the receiver pocket is possible through the use of curved ion-exit slits and accelerating electrodes. Preliminary testing of this concept in actual separations demonstrated its feasibility and equipment conversion in this system is expected to reduce pocket fabrication costs.

C. Ion Sources

The effectiveness with which isotopes are separated is largely dependent upon ion source performance. Energy spread, z-wise divergence, sparking,

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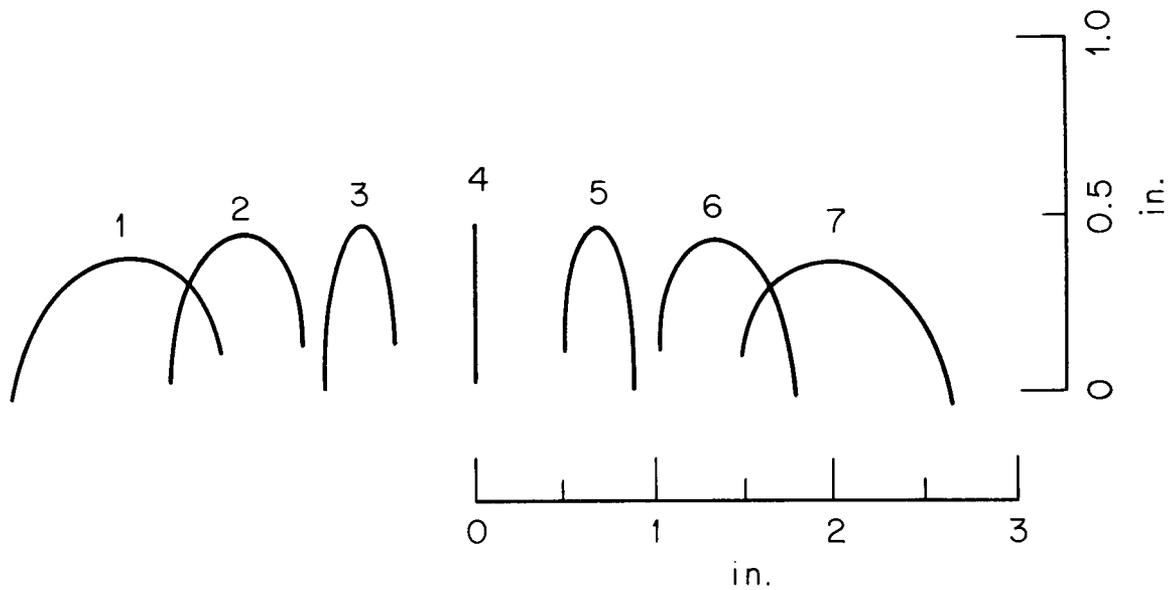


Fig. I-7. Focal Patterns of Specific Ion Trajectories Outlining the Composite Beam Pattern Expected from "Football" Shims. Initial Angle -10° to $+10^\circ$.

<u>Curve</u>	<u>Z, cm</u>	<u>Curve</u>	<u>Z, cm</u>
1	-6	5	2
2	-4	6	4
3	-2	7	6
4	0		

and output modulation all contribute to isotopic contamination. Efforts to minimize these effects and provide increased ion output are an integral part of the development program.

Sputtering Source

For satisfactory calutron processing, charge feeds must be relatively volatile and stable at elevated temperatures. These characteristics are lacking in compounds of the platinum metals group, and vaporization of the element (used as feed material in separating these isotopes) is accomplished by electron bombardment heating.

Electron bombardment sources are difficult to construct, and in operation they yield only a fraction of the output normally realized during operation of conventional sources. In order to develop an alternate method for charge transport, several experimental models of sputtering-type sources were constructed and tested. Metallic copper, used as the charge material, was sputtered into the arc region at an acceptable rate of 1-5 g/hr. The problem of maintaining the integrity of insulators in the presence of so much copper was solved, but the ionization of sputtered copper proceeded at such a slow rate that practical ion outputs could not be achieved from the concepts tested.

Miniature Sources

An increasing number of projects require the use of pre-enriched feeds where an increase in total recovery and process efficiency is especially desirable. Miniaturization of ion sources is an approach to this immediate problem, and the sources themselves could be applied directly into a sector instrument whenever the sector program is finalized.

Two sources of reduced size (one-half and one-quarter "standard" size) have been designed, constructed, and tested in gas separations. Ion outputs equal to at least half those received from standard units have been achieved in a majority of test runs.

Each source is mounted on a regular size faceplate and insulator to minimize construction costs during initial testing of these new concepts. The major components of these sources are shown in exploded view in Figs. I-8 and I-9. The first and larger of the two is similar in concept to the standard high temperature source developed in 1957-58. The second is considerably less complex and employs the concept of slip-fits to simplify assembly and make remote servicing possible. The assembled units ready for placement in the supporting subassembly (Fig. I-10) are shown in Fig. I-11. The larger unit shown in Fig. I-11 for comparative purposes is the source assembly considered to be of standard size.

Each source was operable in its original design, and only slight modification of the drain system has been incorporated in the quarter-scale model in an effort to improve its operational characteristics. These

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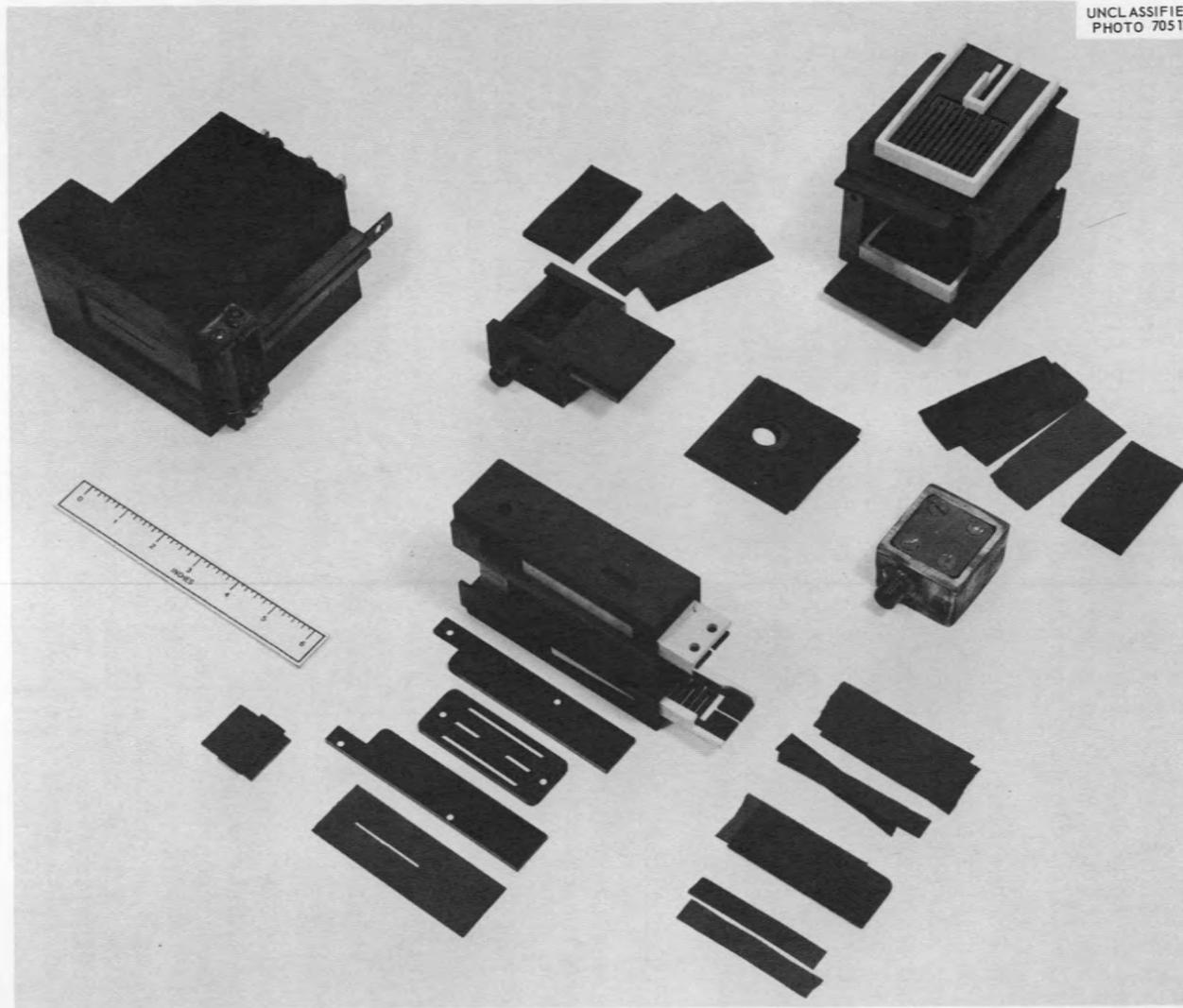


Fig. I-8. Components of the "Half-Scale" Ion Source (Oven-Arc Assembly).

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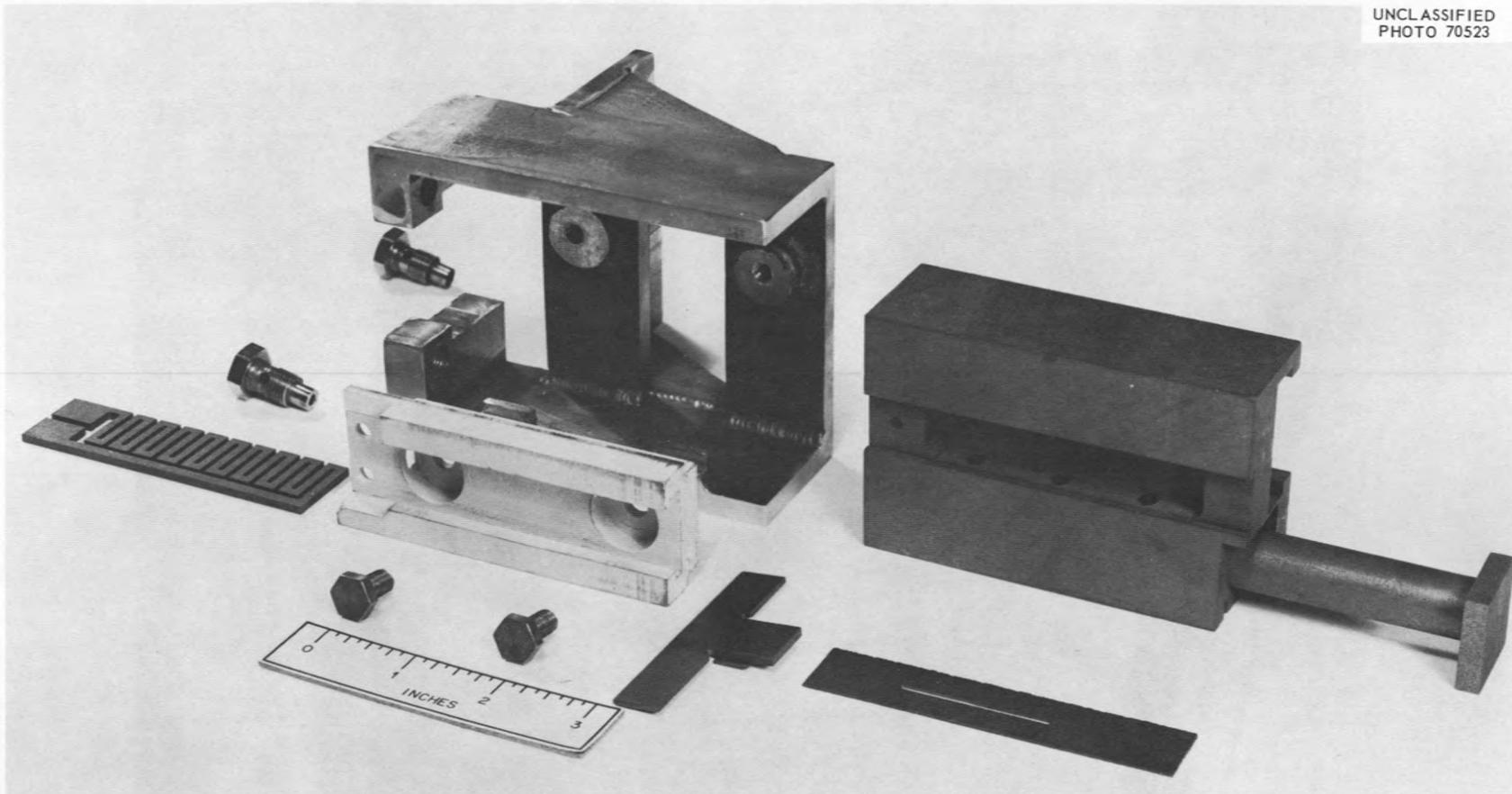


Fig. I-9. Components of the "Quarter-Scale" Ion Source (Oven-Arc Assembly).

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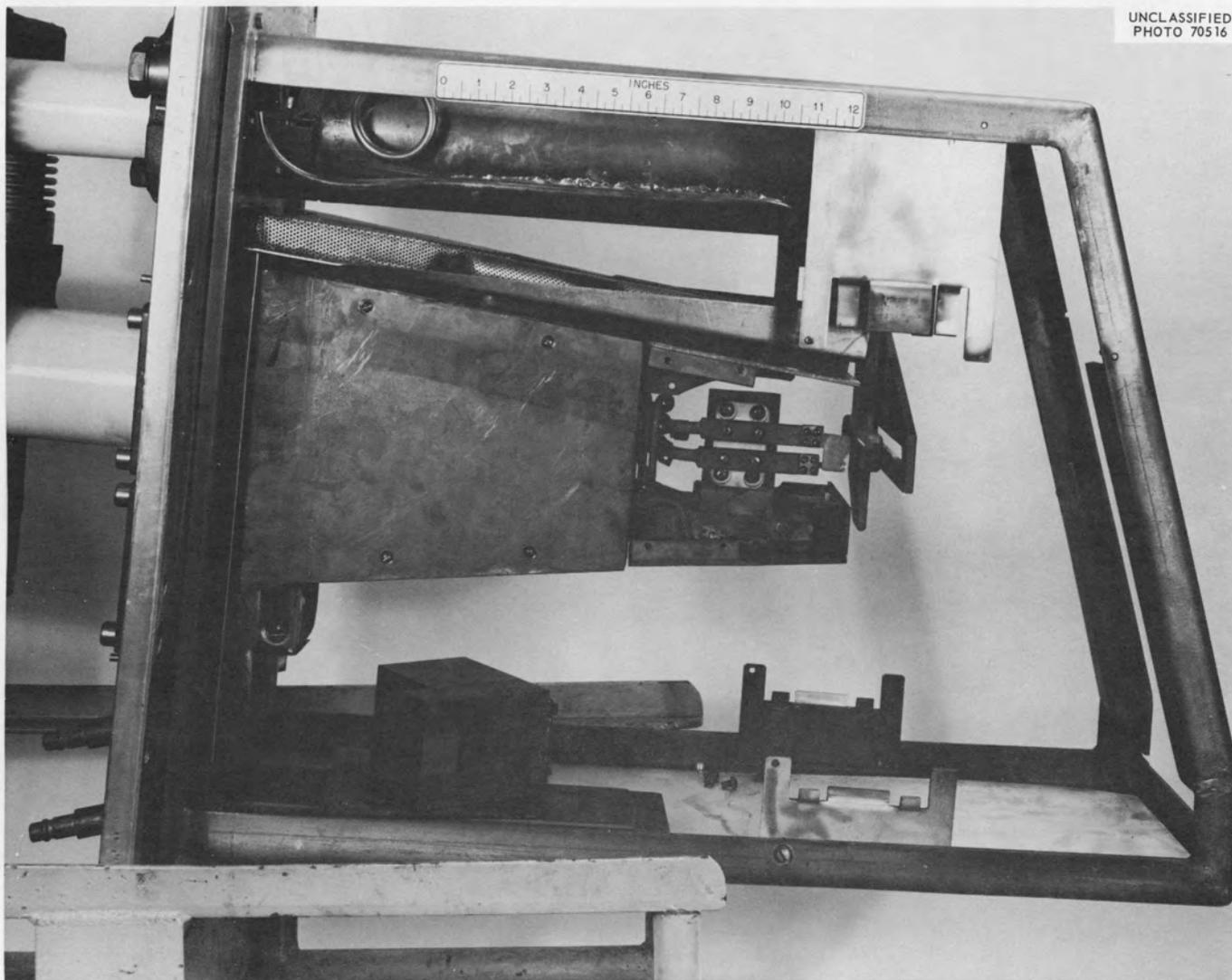


Fig. I-10. Supporting Subassembly for Miniaturized Sources.

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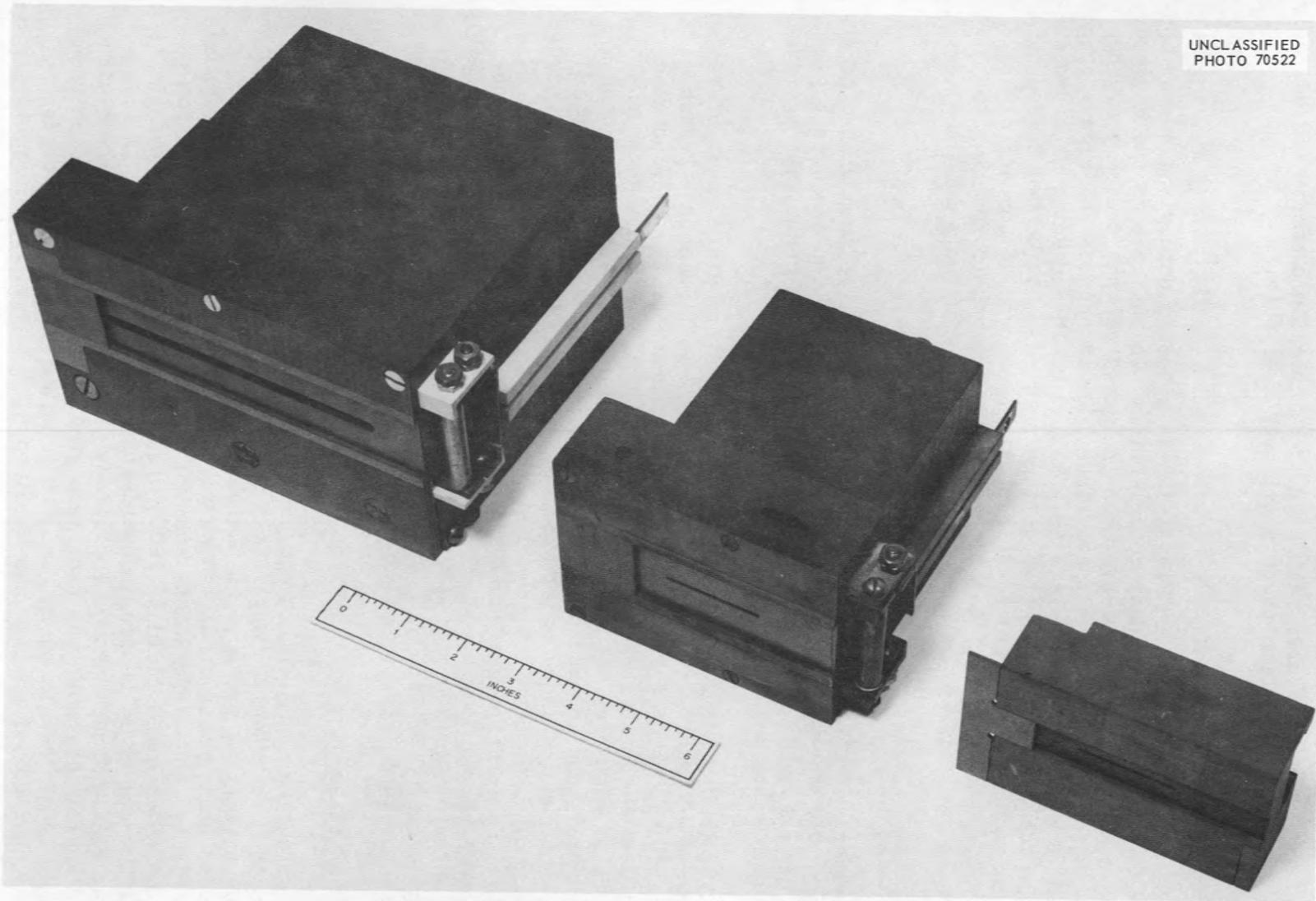


Fig. I-11. Oven-Arc Assemblies (Standard, "Half-Scale", and "Quarter-Scale").

sources are being used as an integral part of the source study program since they permit the observation of arcs formed in chambers of radically different dimensions from those previously available.

Sector-Type Ion Source

The source concept given in Fig. I-12 is proposed solely for sector application, and the unit is under construction. In this design the diameter of the supporting insulator has been increased so that source charging can be accomplished through the airlock located outside the high voltage plate. This high voltage plate is nonexistent in conventional sources, and for standard units all high voltage services must be fed through a 2-in. diameter opening provided in the insulator. The concept of a high voltage plate simplifies construction problems in addition to making charge loading possible without removing the unit from the tank.

Two-Arc Source

Improvement in total ion output is sometimes possible through use of a two-arc system. This approach has limited application since isotopic purities are generally lowered and some complex ion beams make the use of two arcs impractical.

Recent requests for tungsten isotopes at an intermediate level of isotopic purity made use of a two-arc source of the M-16 type appear feasible. As initially designed for internal feeds, these sources incorporated two discrete charge ovens and arc chambers and provided independent control of each arc. Since tungsten will be internally chlorinated and arc control appears less critical whenever internal chlorination is used, this source is being altered to provide a single oven. One source of this type has been fabricated and subjected to test runs using WO_3 charge. Performance was satisfactory and maintenance and equipment costs can be reduced through elimination of one oven and heater assembly. The isotopic purity achieved meets separation specifications even in the presence of WO^+ and WCl^+ sideband beams from the lower arc which strike the faceplate of the top collector assembly.

Enclosed Filament

When observing the oscillations present in a collected ion beam and attempting to establish a correlation between them and source conditions, it was noted that pressure in the collimating slot-filament region affected beam quality. An ion source was modified to completely encase the filament in a vapor-tight enclosure provided with a connection through which gases could be fed. Simultaneously a gas (either the same or different) could be fed independently into the conventional collimating slot and arc chamber locations. When test runs were made with argon and/or nitrogen, the best beam quality was always achieved when a majority of the gas being used was fed into the filament enclosure. Better than normal quality was attained with 1-2 ma beam currents whenever a high degree of one gas (support) was fed into the filament region

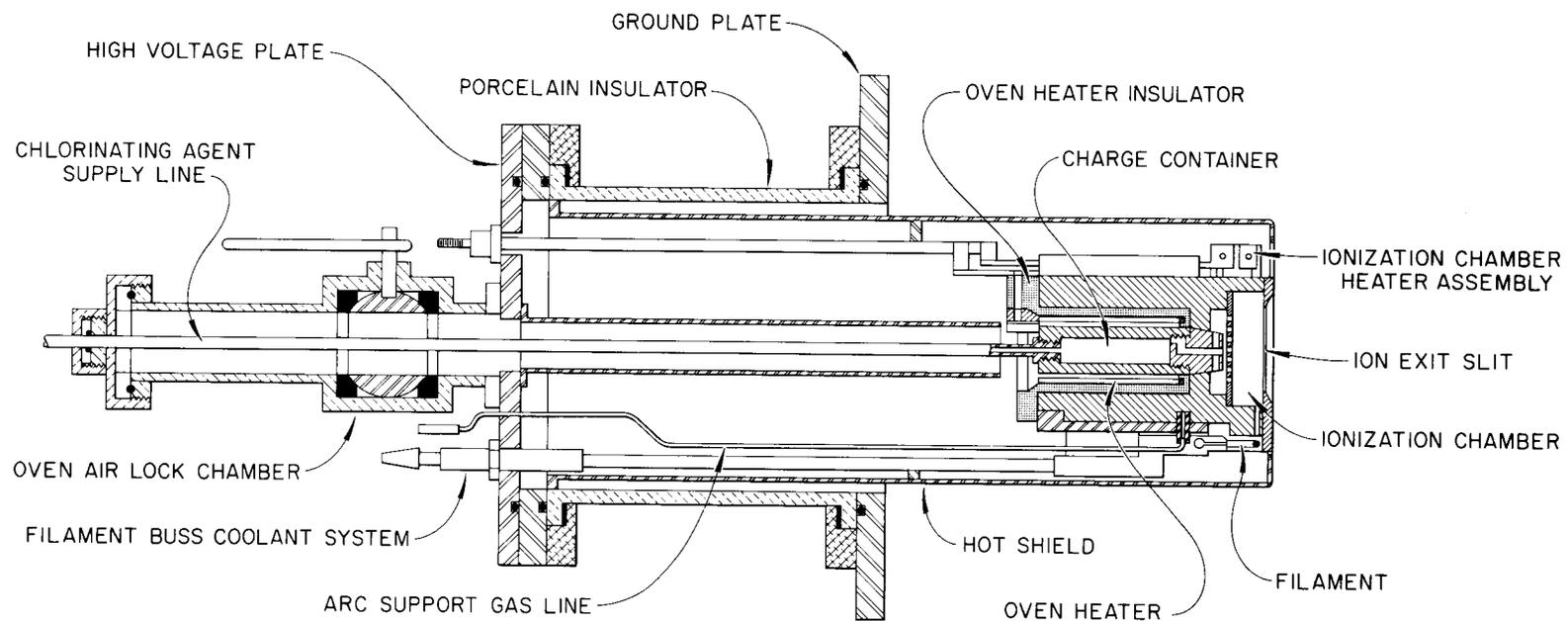


Fig. I-12. Sector Type Ion Source.

and only small quantities of charge gas were admitted into the arc chamber. This action suggests that support gas permits an arc to be struck between the filament and collimating slot to provide stability, and subsequent admission of charge gas permits the major portion of the arc to be formed at much lower pressure.

The application of these observations into separations involving large quantities of solid charges is uncertain; but in areas such as second-pass operations, this technique could provide a means of realizing improved process efficiency and increased isotopic purity.

Electrode Geometry Considerations

Separator performance is influenced by the extent to which the ion beam is collimated by the electrode system. Studies show that electrode heating and ion losses occur whenever electrode offsets are not optimized.

Present sources provide for in-run adjustment of both negative and grounded electrode offsets relative to the source, but the movement is such that the two electrodes bear a fixed relationship to each other. The validity of using such an adjustment is being examined both by direct experimentation and by calculation of approximate particle trajectory under these conditions.

In determining electrode geometry of ion sources for use in standard calutrons as well as ion sources to be used in other types of separators, it is necessary to accurately offset the electrodes in order to ensure minimum power supply drains and unnecessary loss of ion output. The offset is in a direction normal to the electric and magnetic field vectors, and is equal to the amount of perpendicular displacement experienced by a charged particle in passing through the electrode system.

The following derivations are applicable in approximation of electrode offsets for sources with ion extraction perpendicular to a magnetic field. The treatment assumes a uniform field between electrodes and ignores the effect of electrode thickness. Particles are assumed to originate at the center of the coordinate frame with an initial velocity of zero. Figure I-13 illustrates the coordinate system, field orientation, and electrode arrangement used in the development of the equations of motion for a charged particle moving in an $E \times B$ field.

The time required for a particle to traverse the distance d_1 between electrodes 1 and 2 is given by

$$t_1 = \frac{m}{Bq} \cos^{-1} \left(1 - q \frac{B^2 d_1}{E_1 m} \right). \quad (1)$$

The displacement of the particle in the y direction in Region 1 as a function of time is given by

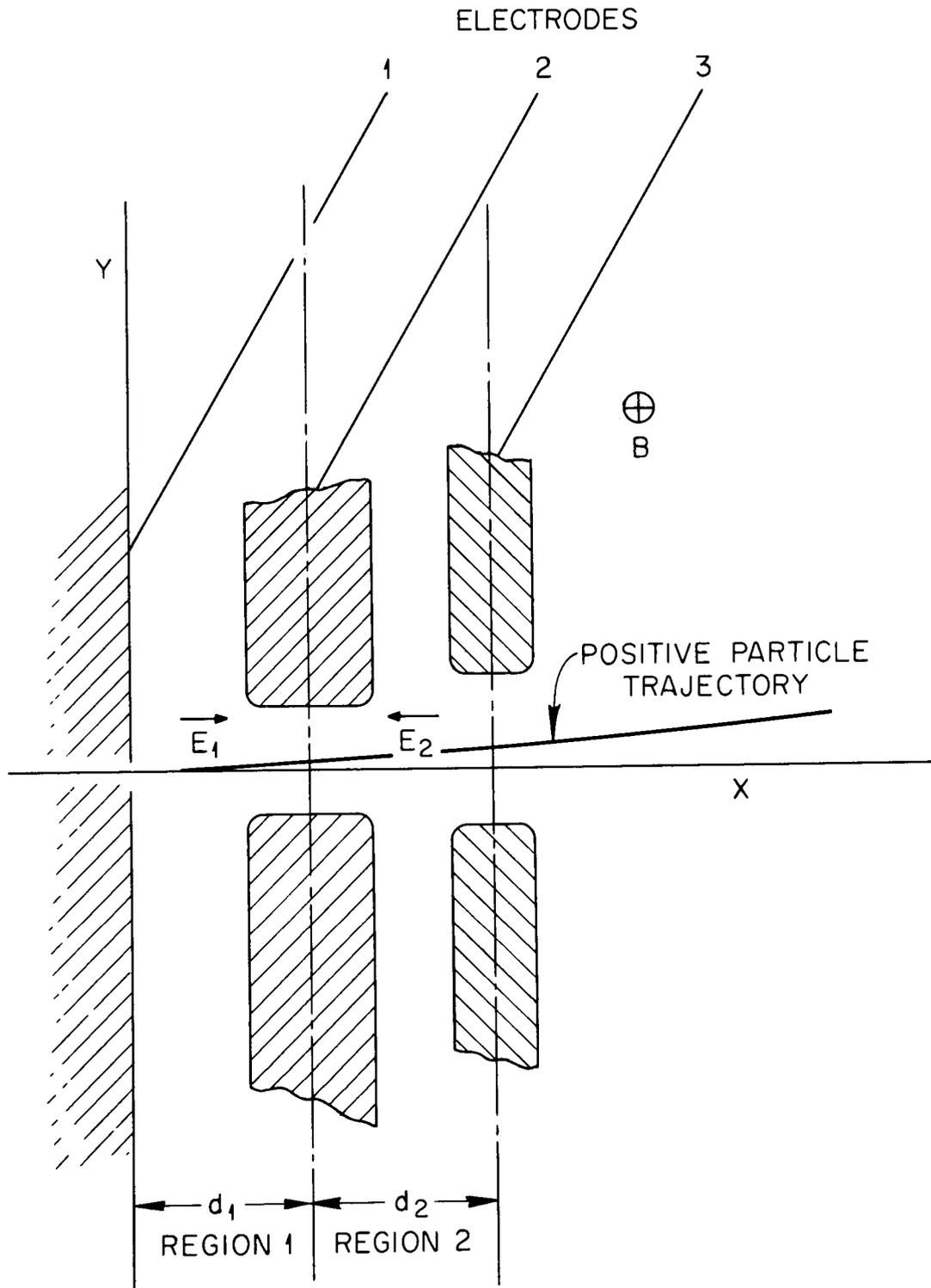
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Fig. I-13. Graphical Representation of Concepts Used in Electrode Offset Studies.

$$y_1 = \frac{E_1}{B} t_1 - \frac{E_1}{q} \frac{m}{B^2} \sin \frac{Bq}{m} t_1, \quad (2)$$

while the velocity component in the y direction in Region 1 may be written

$$y_1' = \frac{E_1}{B} (1 - \cos \frac{Bq}{m} t_1). \quad (3)$$

In this instance the value y_1 corresponds to the offset required between electrodes 1 and 2.

Similarly the equations for the displacement of the particle in the x direction of Region 1 and its velocity component in that direction were developed,

$$x_1 = \frac{E_1 m}{qB^2} (1 - \cos \frac{Bq}{m} t_1), \text{ and} \quad (4)$$

$$x_1' = \frac{E_1}{B} \sin \frac{Bq}{m} t_1. \quad (5)$$

After traveling the distance d_1 the particle moves into Region 2 where the influence of a deceleration field affects the particle trajectory. At time of entry into Region 2 the particle has been displaced a distance y_0 and travels with components of velocity x_0' and y_0' in the respective directions. The equation of position in Region 2 in the x direction may be written

$$x_2 = \left(\frac{E_2 m}{qB^2} + \frac{my_0'}{Bq} \right) \cos \frac{Bq}{m} t_2 + \frac{x_0' m}{Bq} \sin \frac{Bq}{m} t_2 - \frac{E_2 m}{qB^2} - \frac{my_0'}{Bq} + d_1, \quad (6)$$

and the equation for the velocity component in the x direction is

$$x_2' = \left(-\frac{E_2}{B} - y_0' \right) \sin \frac{Bq}{m} t_2 + x_0' \cos \frac{Bq}{m} t_2. \quad (7)$$

The position equation and velocity component equation in the y direction for Region 2 may be written

$$y_2 = \left(\frac{E_2 m}{qB^2} + \frac{my_0'}{Bq} \right) \sin \frac{Bq}{m} t_2 - \frac{m}{Bq} x_0' \cos \frac{Bq}{m} t_2 - \frac{E_2}{B} t_2 + y_0 + \frac{m}{Bq} x_0', \quad (8)$$

$$y_2' = \left(\frac{E_2}{B} + y_0' \right) \cos \frac{Bq}{m} t_2 + x_0' \sin \frac{Bq}{m} t_2 - \frac{E_2}{B}. \quad (9)$$

To complete the displacement in the y_2 direction for the offset of electrode 3 the value of t_2 should be calculated from Eq. 6 when $x_2 = d_2$.

This value when substituted into Eq. 8 will yield the proper offset for electrode 3 in relation to electrode 1.

NOTE: t_1 = time of flight in Region 1
 t_2 = time of flight in Region 2
 m = mass of particle
 B = magnetic field strength
 q = charge of particle
 E_1 = electric field strength in Region 1
 E_2 = electric field strength in Region 2
 d_1 = electrode gap distance between electrodes 1 and 2
 d_2 = electrode gap distance between electrodes 2 and 3
 y_1 = displacement of particle in y direction in Region 1
 y_1' = velocity component of particle in y direction in Region 1
 x_1 = displacement in x direction of particle in Region 1
 x_1' = velocity component of particle in x direction in Region 1
 x_2 = displacement in x direction of particle in Region 2
 x_2' = velocity component of particle in x direction in Region 2
 y_2 = displacement in y direction of particle in Region 2
 y_2' = velocity component of particle in y direction in Region 2
 y_0 = displacement of particle in y direction upon entrance into Region 2 from Region 1
 y_0' = velocity component of particle in y direction upon entrance into Region 2 from Region 1
 x_0' = velocity component in x direction of particle upon entrance into Region 2 from Region 1.

D. Ion Receivers

Calutron isotope separation involves the collection of 35-keV ions in suitable receiving pockets. Space limitations imposed by beam dispersion limit collector pocket size to a box-like structure $\sim 3/8 \times 8 \times 3-1/2$ in. Ions saturate the surface covered by direct impact within minutes; and, when this equilibrium level is reached, the deposited isotopic material as well as the collector pocket is sputtered away by the incoming ion beam. In general ~ 5 atoms of pocket material are dislodged by each ion entering the collector. This value varies (both with material and ionic species) but in most separations, isotopic material of solid form is retained primarily by trapping sputtered particles. Both the placement of the pocket and its physical dimensions are effective in increasing isotopic retention.

Saturation and Penetration

In collection of isotopes having very low natural abundance (i.e. Ca^{46} which is 0.003% of the normal element), advancement has resulted from collecting separated material in the surface actually struck by the high energy ions. Here penetration is of the order of 100 atomic diameters, and the surface saturates at 10-100 $\mu\text{g}/\text{cm}^2$.

Recent work with thorium ions striking both normal and pyrolytic types of graphite show typical quantities trapped in the target material (Table I-2); Fig. I-14 shows the penetration profile. The equipment arrangement used for these experiments provided for continuous rotation of up to 54 sample specimens per run and produced statistically reliable data. Figure I-15 shows a similar receiver used in calcium ion collections to study the saturation value for Ca^{46} on various materials and to determine the sputtering ratio of graphite by Ca^{40} at 38 kev. Various target materials are listed in Table I-3 with values retained and isotopic purities achieved.

Sideband Beam Production

Extended operation with SiS_2 charge where both the Si^+ and S^+ ions are being monitored shows a striking degree of correlation between the ratio of beam currents (Si^+/S^+) and the temperature of the charge oven. Further investigation with chromium and chlorine shows a similar but less pronounced effect. In both instances the ion current of the non-metallic charge constituent is increased whenever the operational temperature is lowered. Readjustment of arc voltage and current will not restore the original current relationships. The ability to exercise a certain degree of control over sideband beam currents is desirable. The concept that control is possible through pressure regulation could mean realization of improvements in operational efficiency which would be particularly applicable in second-pass separations.

E. Charge Evaluation

In early calutron separations the choice of a feed compound for ionization was limited to elements or compounds which could be vaporized in sources having a temperature range of 300 to 600°C. Source development increased the temperature range and thus allowed more choice in feed materials. It became evident that operations could be altered tremendously by the charge material used.

Internal Chlorination

The technique of preparing volatile feeds by chemical reaction (chlorination or oxidation) within the ion source has exerted a greater influence on operations than any other single phase of charge preparation or choice. Alteration of feed containers makes possible the internal chlorination of ~50% of the elements processed. Experimentation in choice and form of the material to be chlorinated (oxide, carbonate, sulfide, or metal) reveals a general preference for oxides or carbonates that have been formed at low temperatures. Compounds formed at high temperatures require the use of more chlorinating agent (CCl_4) per unit time and provide less ion output than do the low-fired compounds.

The internal chlorination of a mixture of tantalum and hafnium oxides resulted in an outstanding improvement in tantalum operation. Increased

Table I-2. Summary of thorium deposition in graphite

Bombardment energy, kev	Bombardment time, hr	Average integrated current reading, ma	Type of graphite	Quantity of Th deposited, $\mu\text{g}/\text{cm}^2$	Actual quantity of Th embedded, $\mu\text{g}/\text{cm}^2$	Method of analysis	Collection efficiency, %
40	5	0.020	pyrolytic	10	8.76	activation	88
40	5	0.020	impregnated	10	10.28	activation	100
36	8	0.500	C-18	385	65.6	colorimetric	17

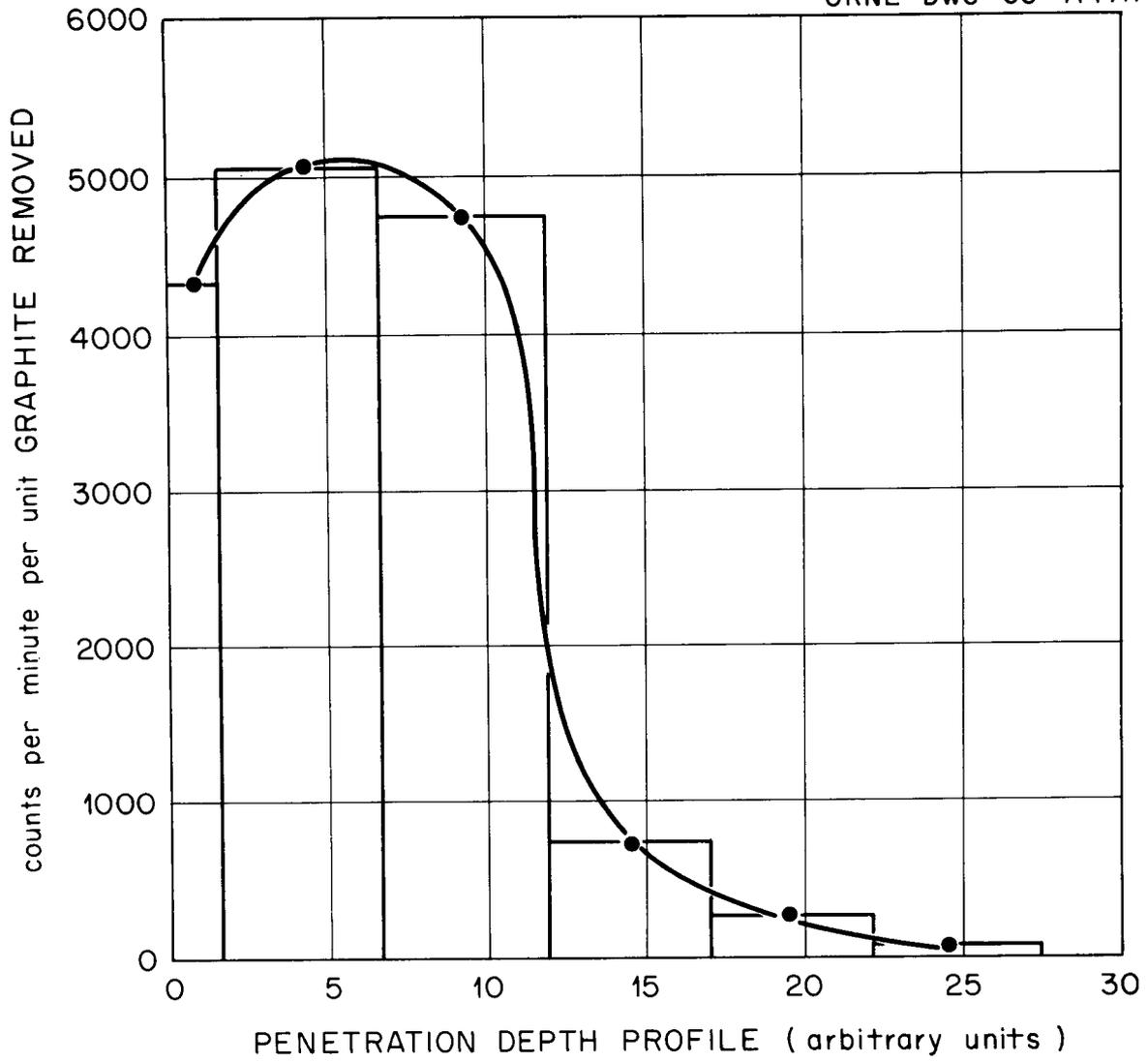
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Fig. I-14. Penetration Depth Profile.

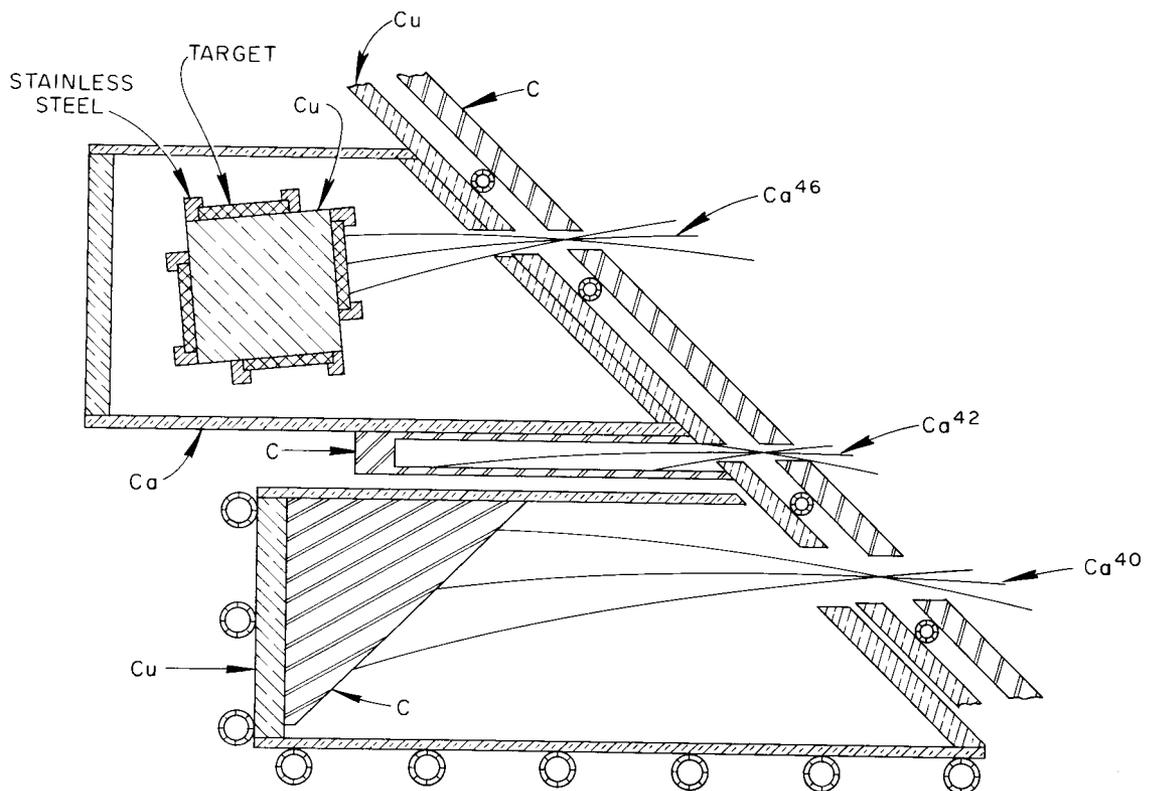
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Fig. I-15. Cross Section of Receiver Used in Sputtering and Saturation Studies.

Table I-3. Bombardment of targets with Ca^{46} ions using rotational receiver

Sample number	Target material	Estimated wt Ca^{46} , mg	Recovered wt Ca^{46} , mg	Retention in target, %	Assay Ca^{46} , %	Ratio $\text{Ca}^{46}/\text{Ca}^{44}$
1	normal Cu	0.840	0.335	40	42.0	17.4
1	Cu crystal 110*	0.840	0.385	46	42.5	17.9
1	Cu crystal 112*	0.840	0.357	42	40.8	18.1
1	C-18 graphite	0.840	0.502	60	36.1	14.7
2	normal Cu	0.960	0.210	22	45.6	15.7
2	Cu crystal 110*	0.960	0.350	36	47.0	18.2
2	Cu crystal 112*	0.960	0.375	39	45.6	14.8
2	C-18 graphite	0.960	0.525	55	33.0	12.7
3	normal Cu	1.100	0.355	32.3	47.7	18.8
3	Cu crystal 110*	1.100	0.413	37.5	50.0	19.92
3	Cu crystal 112*	1.100	0.255	23.1	49.3	19.4
3	laminated C	1.100	0.450	41	41.8	17.6

* represents the crystalline plane normal to the ion beam

NOTE: average parameters: ion energy - 37.5 kev; target area - 15 cm^2 ; ion current - 0.22 $\mu\text{a}/\text{cm}^2$

ion output was achieved, and the presence of Hf^{180} in the beam permitted monitoring of the current in the Ta^{180} collector. (Natural abundance of Ta^{180} is 0.012%, and ion current of this isotope alone is too small for accurate monitoring.) Assays of the samples collected showed the Ta^{180} content to be 0.6% which is almost a 70% increase over previously collected single-pass material.

During the year charges for Cd, Ta, Yb, Cr, Pu, and Rb separations were prepared by the internal chlorination process. Exploratory work has shown the desirability of extending this technique to planned separations of W and U. No improvement in operation was noted when internal chlorination was applied in separations of Ni, Fe, Pb, or Hg.

Silicon Sulfide

Another significant improvement in operations resulted when silicon sulfide was substituted for silicon tetrachloride, previously the preferred compound. Through use of SiS_2 feed, the ion output of Si^+ was tripled and S^+ was collected simultaneously with the Si^+ at essentially the same rate as expected in a normal sulfur separation. At the same time, appreciable increases in the isotopic purity of Si^{29} and Si^{30} were achieved. The results, summarized for approximately nine months of operation, are shown in Table I-4.

Table I-4. Ion outputs and isotopic purities achieved for silicon using SiS_2 and SiCl_4 feeds

Charge feed	Ion output, ma		Isotopic purity, %	
	Peak	Average	Si^{29}	Si^{30}
SiS_2	162	71.6	>95	>95
SiCl_4	45	26.8	88	90.2

F. Contamination

Attempts to improve isotopic purity are often futile because of the large number and complexity of factors contributing to contamination of collected samples. Effects of pressure scattering, charge exchange, residual background pressure, and even collector pocket impurities are recognized as being detrimental to sample purity. Contamination is also influenced by undetermined processes and the combined action of all factors makes precise prediction of isotopic purity almost impossible.

Prediction of sample purity using electrical resolution (beam scanning) results in an estimate that is too high. Application of a "contamination curve" derived from past separation data and adjusted for isotopic abundances permits a realistic estimate for the sample produced from any feed composition if current performance is equal to past performance.

The use of oscilloscope monitoring of a collected ion beam has revealed an unexpected degree of beam oscillation existing throughout a separation run, and it is evident that oscillatory conditions contribute to contamination.

Contamination Curve

For a long time the enhancement formula was the only relationship used for measuring the efficiency of separating isotopes in the calutron. This formula,

$$\eta = \frac{\frac{cp}{1 - cp}}{\frac{cf}{1 - cf}},$$

where

cp = concentration of wanted isotope in the product,

cf = concentration of wanted isotope in the feed,

was limited in use to comparison of like separations performed at different times. It was not possible to relate the efficiency of separating isotopes of different elements, separating different isotopes of the same element, or separating the same isotope if the feed abundance changed (e.g. in second-pass separations).

In many other methods of separating isotopes, the above formula tends toward some ideal limit (e.g. $\sqrt{\frac{m_1}{m_2}}$ where m = mass). Under idealized conditions in the calutron process, the isotopes should be completely separated. This does not occur in practice since a number of different factors prevent complete separation and lead to contamination of the collected isotopes by small amounts of other isotopes, particularly those closest to the mass under consideration.

Another method of examining the separating efficiency of calutrons has been developed both at ORNL and in various laboratories in Europe. This method is based on the fact that the beam shape or profile is the same (within experimental limits) for all isotopic beams of an element regardless of their relative size or relative mass position. This uniformity makes it possible to consolidate all useful information in the mass analyses of the various isotopes (except statistical data) into a few constants which express the efficiency of the separation. The process involves "fitting" the contamination in the collector pocket under consideration as a function of the mass difference between the contaminating isotopes and the collected isotope.

Usually data fits between contamination and mass difference are determined graphically. Often contamination from higher mass isotopes is greater than that from lower mass isotopes with the same mass difference, and two fits are required. Empirical study showed that the percentage of normalized contamination was equal to

$$100e^{-k\left(\frac{|\Delta m|}{m}\right)^{1/4}}$$

for the majority of elements separated. For many reasons (e.g. peculiar point distribution, non-random nature of some of the deviations, and insufficient number of isotopes in some elements) data are too few and too inferior to permit determination of all three constants (100, k, and $1/4$) in the above equation. In the majority of separations, the fixing of two constants permits a reasonable data fit.

There have been only a few exceptions to this data fit, usually caused by known properties of specific elements. Mercury and sulfur have a relatively low vapor pressure under normal conditions, but it is high enough to cause extra contamination in the calutron. In separations of these two elements, contamination is usually constant for all isotopes. In separations for some other elements, neutral contamination is low enough that a combination of the function given above and a constant are required to define the mass-to-contamination relationship.

With the platinum and palladium group of elements, the vapor pressure of calutron feeds is so low that special methods (e.g. electron bombardment) are used to get sufficient vapor into the arc sources. In these types of separations, contamination by adjacent isotopes is larger than usual; therefore, it is necessary to determine two constants from the data. The use of $|\Delta m|^{1/4}$, however, still linearizes the data on plots of log contamination vs $|\Delta m|^{1/4}$.

A third exception to the usual data fit developed in the calcium separations. Rather suddenly the separations improved substantially (i.e. the assay of Ca^{46} rose from 30 to nearly 45%). In fitting the new data into the above treatment, it was found that the constant coefficient of 100 had to be reduced to 0.1, and the slope of contamination plotted on semi-log paper using $|\Delta m|^{1/4}$ as the abscissa was reduced (indicating reduction in contamination by adjacent isotopes). The reason for this process improvement has not been definitely identified, although it is probably related to source changes made for the purpose of reducing the chemical reaction between graphite and hot calcium vapor. If this alteration in equipment is responsible for the noted improvement, it has not had a similar effect on separations of other elements.

Excluding these exceptions, this data fit between contamination and mass difference permits consolidation of a 15-year accumulation of calutron data. All pertinent results can be incorporated into a few constants which represent average contamination. In instances where inventory sample analyses are used, the data are indicative of the average performance of several calutrons over many months of operation. Use of preliminary sample analyses presents the average performance of individual calutrons for shorter periods ranging from 50 to 2000 hr.

Theory does not predict how the separation efficiency of the calutron varies with atomic weight of the processed isotopes. Examination of

available analytical data reveals that a semi-log plot of contamination by isotopes differing by one mass unit from the collected isotope against $\frac{1}{|\Delta m|^{1/4}}$ produces approximate linearization of data (excepting the platinum group, mercury, and sulfur). This relationship extends the range of data that can be directly compared and makes it possible to evaluate individual separator performance in any given series against a standard performance value for all past experience for all isotopes of all elements.

On the basis of contamination curves and mass differences, a remarkable consistency in separation performance is demonstrated. The degree of contamination noted appears to vary by less than a factor of 2 between various elements from various separators. Marked reduction in contamination has been noted with introduction of new ion sources and techniques. The most significant recent improvements are shown with Si, Ca, Pu, and those elements processed in the new inhomogeneous field separators.

Oscillatory Conditions

In approaching a study of contamination due to oscillatory beam conditions observed at the collector, it was essential to determine if the oscillations were also observable near the source. Ion receivers located to intercept the beam some 8 to 10 in. from the source received a modulated current, this modulation frequency being related to parts of the frequency spectrum noted at the conventional receiver. Observation of the insulated anode positioned at the anti-filament end of the arc reveals that the same frequency appearing near the source also appears on the anode of the arc chamber from which the beams originate. Phase relationships between the oscillations have not been determined, and definite correlation between the frequency and amplitude of oscillations at the source and the quality of the collected ion beam has not been established.

Oscillatory conditions in the arc chamber can be altered by voltage and current, pressure, location of vapor entry into the region, or dimensions of the arc chamber itself. Strong evidence exists that the region between the collimating slot and the filament is a critical location from which oscillations arise. Adequate control of charge vapor in the collimating slot region has resulted in a relatively high degree of stabilization of beam quality.

In addition to frequencies observed in the arc region, oscillations are noted at the collector and it is possible these oscillations are being generated within the beam. Since some intensity modulation of the beam exists at the source, this action may provide an electron loss mechanism. During periods when the density of positive ions in the beam decreases, rapid electron loss could occur which could be more rapid than the replacement rate. In this situation, unstable conditions could persist.

In order to investigate these electron losses, the 255° separator was fitted with an arrangement of screens and plates bounding the beam. A screen having $\sim 1/8$ in. mesh was placed on each side of the beam and was

maintained at ground potential. Solid plates were positioned $\sim 1/2$ in. back of the screens and were supported by insulators capable of biasing the system to >300 v. A second set of plates positioned $1/2$ in. from the bias plates was provided at ground potential. Electrons could supposedly penetrate the screen and be repelled by the plate carrying the bias voltage. Experiments performed under normal operating conditions failed to provide definite conclusions relative to the effect of electron loss on beam quality. The application of negative bias up to 300 v failed to produce any noticeable improvement in beam quality in the 255° separator system.

Measurements of current striking the plates seem to indicate that essentially equal numbers of electrons and ions leave the region and strike an unbiased plate; most of the electrons leaving the region have energies in the direction of the field of <25 ev; and ions leaving the region have a broader energy spectrum than do the electrons. Application of positive bias to the plates increased the electron loss rate and produced an extreme beam blowup. It is concluded that beam instability as noted at the receiver is produced by mechanisms more complex than electron losses resulting from fluctuation in positive ion density.

Spectrographic Beam Analysis

Spectrographic examination of the calutron beam in the region of the ion source and receiver is being undertaken to aid in understanding the processes of isotopic contamination. An ARL-Dietert reflection grating spectrograph is being used to record the spectra of a calcium ion beam (Fig. I-16).

Calcium has a relatively simple spectrum, and ion or atom velocities can be determined from observed Doppler shifts in spectrum lines. Energies, in turn, can be calculated from mass and velocity relationships. De-excitation time is considered to be $\sim 10^{-8}$ sec which is so short a time interval that observed phenomena are known to occur at a distance of less than 0.5 cm from the point of observation.

The ion beam as observed in the electrodes of the source is shown to contain ions having an energy distribution up to the sum of the applied electrode potentials. These are the singly-charged ions intended for eventual collection. Also present (in yet undetermined percentages) are singly-charged ions having almost twice the energy desired for collection. An appreciable quantity of these ions result from charge transfer associated with an initially doubly-charged ion and neutral calcium atoms. Neutral atoms of calcium are also abundant at the receiver. Most of these neutrals have thermal energy but a small fraction has energy equal to that of the collected ions. In addition to the 35-kev ions being collected, ions of almost zero energy are present in the received beam.

The observed presence of a wide energy spread in the ions composing the beam being collected substantiates certain concepts of contamination

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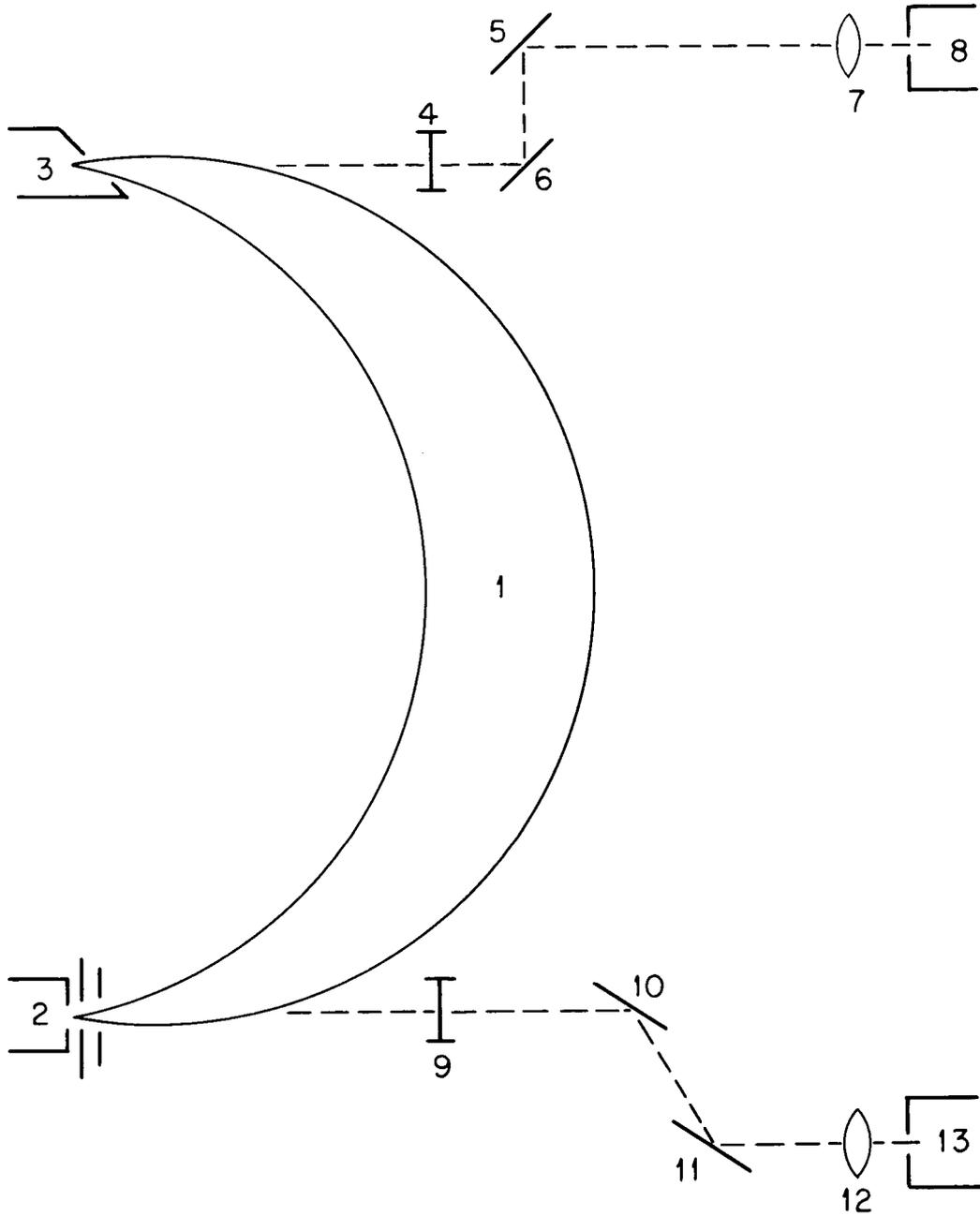


Fig. I-16. Positioning of Spectrometer at Calutron Viewing Windows.

Beam	1	Receiver	4	Front Silvered Mirror	5,6,10,11
Source	2	Source Window	9	Spectrograph	8,13

developed through the use of statistical data analysis and plotting of contamination curves. Present experimental techniques are inadequate to permit quantitative determination of beam components, and other refinements are planned which will permit the study of beams other than calcium.

Mercury Separation

Satisfactory enrichments of mercury isotopes in past separation series have been achieved only when costly refrigeration of the calutron liner was employed. In order to obtain the quantities and enrichments of mercury isotopes required in a currently scheduled separation, eight separators must be used for a period of at least one year. Refrigeration of eight calutron liners over such a long period would increase the cost of such a series to an almost prohibitive level.

The possibility of solving this problem of neutral mercury vapor contamination by less costly methods is being explored. Approaches have been from several viewpoints: (1) reduction of neutral mercury background pressure in the calutron by improvement of control over charge feed rate and isolation of the source from the collector region to provide differential pumping from each region; (2) separation of neutral mercury contamination from the enriched sample by distillation techniques incorporated into the recovery process.

An internally-valved charge feed system has been developed and its use in mercury separations has demonstrated satisfactory feed control and beam resolution. Tests show that this feed system yields 30% higher maximum total ion output and 40% higher average total ion output than any other system used in previous mercury separations.

Reduction in neutral mercury background by differential pumping on the source and collector regions is planned. The ion beam cross section at the 90° position in calutrons and at the 128° position in the 255° separator prevents total isolation of the source and collector regions. Sufficient isolation of the two regions to provide effective differential pumping can be achieved only in the 255° separator since the focal properties of the calutrons require use of a 90° baffle opening having 10 times the area of the 128° baffle opening used in the 255° separator. Modification of a 255° system to permit this isolation has been completed but has not yet been tested.

In past mercury separations, retention of collected material by amalgamation in silver pockets was only 40.5%. These separations had been performed on the premise that collection of lowered ion currents would lower collector pocket temperatures and these reduced temperatures would increase retention. Recent calculations predict that reducing the pocket temperature by 100°C can reduce the transport of collected mercury out of the pocket by a factor of 3, but at the same time can increase the pumping speed of neutral mercury into the pocket by a factor of 5. Experimental runs in the 255° separator have averaged a 30% increase in ion beam, yet retentions of 63.1 to 88.2% have been achieved. Increases

in collector pocket temperature are thus shown to have increased retention, presumably through better amalgamation at higher temperatures.

G. System Modifications

Electrical

Existing equipment is altered or replaced with improved designs whenever such alteration will improve the process or maintain equal process levels at a substantial reduction in maintenance costs. An example of this approach is the upgrading of magnet regulation which has been accomplished in both XAX and the track magnets by the installation of amplidynes and associated switch gear. In XAX the regulation of the magnetic field is now 1 part in 10,000 compared with the 1 part in 4000 previously attainable. This amplidyne system, in addition to improving field regulation, provides greater versatility in switching generator sets and requires less maintenance effort to keep the facility functioning properly.

Additional effort involving magnetic fields includes the installation of a 400 kw motor generator set to provide magnet excitation and the reconnection of magnet coils in the special separations track to compensate for field losses from the 90-ton yoke to the unexcited system adjacent to the yoke. The change increased the current-carrying capacity of the end coil and made excitation of unused magnets no longer necessary.

Failure of the generator providing XAX magnet excitation current required that the generator unit be removed, and a four-month period was needed to complete necessary repairs. To avoid downtime of the calutrons, a temporary power supply was made of six, 10-v, 700-amp, copper oxide rectifier units connected in series to furnish 60 v at 700 amp dc. One of these units was powered through an induction voltage regulator which was controlled by a "servo-system" (made from a high gain, error sensing amplifier and a Brown Recorder power amplifier with suitable motor driven mechanism). By controlling the output of only one of the rectifier units, sufficient power and control was obtained to permit separation of rubidium isotopes. All of the equipment used in this temporary assembly was available within the Department.

Solid state rectifiers installed in the high voltage supply of the two XAX cubicles cost \$3,600 per cubicle. These supercede vacuum tubes having an annual replacement cost of \$6,000 per year. After two years of operation, no replacement costs have been incurred with the solid state rectifier systems. Installation of similar systems in the beta-type power supplies is being undertaken on a limited basis; however, it is recognized that the increase in peak inverse voltage associated with these supplies may produce rectifier damage equal in cost to tube replacement. Similar advancement is being made in conversion of arc supplies (300 v, 10 amp) to the use of silicon rectifiers. At this time 33 systems have been converted and are providing performance superior to tube-type supplies.

Manufacture of the oscillator tube previously used in the cyclotron has ceased, and circuitry changes necessary to accommodate the new replacement tube have been made. These new tubes do not demonstrate the characteristics associated with the previous models, and cyclotron performance with the new tube is as yet substandard.

Cubicle Relocation

Grouping of the four calutron power supplies for XAX and XBX in a single area provides for centralized operation with a 50% reduction in rotating shift personnel (annual saving of 5 man-years). The move required relocation of each supply and was scheduled so that two separators remained operative throughout the change to minimize the amount of calutron downtime. In the case of the XBX supplies, the change also incorporated substitution of standardized and updated cubicles for the two which had been in continuous use since 1943. Further refinements were made toward automation of gas feeds and installation of balanced rectifier circuits in the arc supplies.

Uranium Laboratory and Wash Lines

Enlargement and updating of the uranium processing laboratory provides the space and equipment needed to chemically process the high purity uranium isotopes separated in the new doubly-contained area. Program scope includes processing of U^{234} , U^{235} , U^{236} , and U^{238} . Because of the degree of its radioactivity, U^{233} will be processed in the plutonium recovery and refinement system.

The renovation of two obsolete wash lines for calutron units is in process to provide segregation between units used in processing stable isotopes and uranium. The need for dual processing facilities is apparent when even uranium content in parts per billion invalidates some uses of stable isotopes. Most of the scheduled uranium separations will be made using pre-enriched feeds which require recycle and recovery of charge materials. Double wash lines will minimize the possibility of feed dilution during the simultaneous processing of two uranium isotopes.

H. Quality Control

Procedures developed for determining average values of contamination (see Sec. F) resulted in reasonable standards for comparison of deviating results. These deviations have been studied with respect to their source, size, and frequency to provide a guide for research and development activities and to aid in establishing the minimum number of mass analyses required for adequate quality control.

In general, most samples analyzed fall within predictable limits from the accepted standard of process performance. In those results which exhibit excessive contamination, the method of comparison frequently allows identification of the source or type of contamination present

(i.e. from normal material, isotopically enriched material, or even errors in transcription of numbers).

Normal material may enter a sample as a result of collector pocket impurity, background tank pressure, severe beam instability, or chemical impurity in analytical reagents. Isotopic impurities are introduced through such means as imperfect beam focusing, ion scattering, charge exchange, beam instability, sputtering of collected material, mismonitoring, flaking of collected material with subsequent transfer to another pocket, or inadvertent mixing of pockets during receiver disassembly or chemical purification. Observation of a single mass analysis will reveal the presence of contamination and frequently will reveal the general type (normal or isotopic). Establishment of the source of the contaminant and the extent to which the other isotopes may also be contaminated may require observation of data from two or more mass analyses performed on the same group of samples.

A major step in quality control could be achieved by development of more precise monitoring measures during the separation process. Isotopic purity is highly dependent upon the observations and judgments made from metered parameters and oscilloscope traces. Attempts to further develop these approaches are in progress, but even if perfected they could not exert control over the possibility of isotopic contamination by inadvertent mixing in operations outside the separator. Complete control would include both in-process monitoring and subsequent mass analysis of the material in each collector pocket. This last approach would entail complete processing and analyzing of material from ~1000 pockets per year.

A practical approach to control was needed and past data were scrutinized to determine the frequency of abnormal contamination and its effect on final samples. Thousands of mass analyses, each showing the percentage of all other isotopes, were examined by plotting the processed data as contamination curves. These curves revealed past samples which contained undue contamination due to unsuspected mismonitoring, undetected flaking or sputtering, accidental mixing subsequent to calutron separation, and the assignment of erroneous values to particular samples. These circumstances occurred infrequently, but the effect on sample purity could be of sufficient magnitude to limit usefulness of the resulting sample.

Two methods of control are now in use: (1) careful monitoring of the ion beam with respect to both output and quality, and (2) contamination curve analysis of data from a preselected pocket taken from each receiver terminated. In the latter approach the pocket containing the isotope which will be most affected by contamination is removed from each receiver, chemically processed, and the resulting isotopic material is analyzed. Other isotopes collected in the same receiver are assumed to be similarly contaminated, and pockets containing these isotopes are subgrouped prior to chemical processing. Generally four subgroups of each isotope are formed according to expected purity, and the material in each group is

assayed prior to further consolidation into one or more final samples. Should the prediction of purity resulting from contamination curve analysis be in error, this subgrouping limits the spread of contamination to approximately one-fourth of the sample and protects the integrity of the major portion of the material separated.

It is recognized that this method of control is based on arbitrary choice of a control isotope, and the degree of control could be intensified to limit the probability of contamination to less than one-fourth of the sample produced. Analyses necessary to extend the degree of control beyond the present point are costly, and the economies realized through single-isotope analysis and associated subgrouping of other isotopes appear to balance the calculated risk taken in not detecting undue isotopic contamination.

I. Chemistry

Charge Preparation

The final choice of charge feeds is based on achievement of maximum ion output consistent with desired isotopic purity. Several factors--vapor pressure characteristics, stability, affinity for moisture, physical form--exhibit a relationship to output, but this relationship is not sufficiently established to allow prediction of calutron performance.

Charges for the following elements were supplied in kilogram quantities this year: Ca, Si, Yb, Pb, Cd, Br, S, Ba, Rb, Cr, W, and Ni. A total of 127 kg of lead chloride, nickel chloride, and nickel bromide was prepared in anhydrous form. Material for Ca⁴⁰ charge (13.5 kg) was prepared as anhydrous chloride from enriched material, and a 2.7 kg quantity of rubidium chloride was prepared from rubidium iodide using ion exchange in the conversion. Tungsten oxide (14 kg) was prepared from available metal scrap and obsolete components, and tantalum oxide (21 kg) was prepared from burned out filaments accumulated during the past few years.

Charge evaluation studies were performed with the following: a mixture of aluminum sulfide and silicon dioxide, barium fluorosilicate, ruthenium dioxide, tungsten disulfide, tungsten hexachloride, tungsten hexafluoride, tungsten oxydichloride, rubidium carbonate, silicon carbide, silicon disulfide, lead oxide, titanium dioxide (with and without carbon dust), cerous oxide (with and without carbon), tantalum pentoxide, nickel oxide, mixture of tantalum pentoxide and hafnium, tantalum nitride, and samarium oxide.

Recycle and Recovery

Final recovery of rhenium and ytterbium feeds yielded 3488 g of rhenium and 10,890 g of Yb₂O₃. These values represent 94% recovery of rhenium

and 88% recovery of ytterbium. Processing of rubidium is incomplete, and recovery of second-pass feeds of Ta, Sa, and Er is in progress.

Conversion

A technique was developed to convert milligram to gram quantities of isotopically enriched bromine (NaBr) quantitatively into anhydrous elementary bromine. The sodium bromide is oxidized by wet chemistry methods using potassium permanganate and acetic acid. Water is removed with a magnesium perchlorate drying trap, and the bromine is collected in an alcohol-dry ice cooled trap before sealing in a Pyrex break seal.

Refinement

Refinement techniques are developed and applied to provide quantitative recovery of isotopes while reducing chemical impurities to the lower limits of detection by optical spectroscopy. Certain uses of isotopes require a higher degree of chemical purification, and additional refinement steps are taken to provide samples in these instances.

The annual chemistry work load involves recovery of material from 900-1000 receiver pockets of 10-12 elements. Subdivision and grouping of this material requires initial refinement of some 400 batches to a chemical purification sufficient for mass analysis. Further consolidation results in some 60-80 inventory samples which are highly refined prior to sampling for chemical and isotopic purity.

Calcium-48 Fluoride Crystal

Initial refinement of Ca^{48} to be used in the formation of a single crystal of Ca^{48}F_2 for Brookhaven National Laboratory was performed by the established method. The resulting crystal was found to contain 0.027 ppm uranium (compared with 0.012 ppm uranium in similar Ca^{48} inventory samples). This level of uranium contamination was objectionable and the crystal was returned to ORNL for further purification. Extension of existing techniques (oxalate and carbonate precipitation) produced a 14.76 g Ca^{48} sample having a uranium content of 0.00046 ppm. Similar treatment of a 40 g sample of Ca^{40} to be used in development procedures prior to growing a second Ca^{48}F_2 crystal reduced its uranium content to 0.00025 ppm. Both samples were sent to Harshaw Chemical Company for preparation of crystals.

II. ELECTROMAGNETIC SEPARATIONS

Nine different elements--Ca, Si, S, Cr, Ni, Rb, Ba, Pb, Cd--were processed in 28 calutrons assigned to the separation process. Tank time available totaled 52,035 hr, and 78.8% of these hours were innage hours.

Table II-1. Summary of current isotope separations

Element	Series	Process efficiency, %*	Charge material	Weight equivalent to monitored ion currents, g**	Number of collector pockets to chemistry*	
Ca***	LO	24.7	Ca metal	Ca ⁴⁸	17.	58
				Ca ⁴⁶	0.316	
				Ca ⁴⁴	177.	
				Ca ⁴³	12.	
				Ca ⁴²	58.	
				Ca ⁴⁰	12,143.	
				<u>12,407.316</u>		
Si***	LW [⊕]	4.6	SiS ₂ +Si	Si ³⁰	49.	28
				Si ²⁹	72.	
				Si ²⁸	1,355.	
				<u>1,476.</u>		
S***	LW [⊕]	1.1	SiS ₂ +Si	S ³⁶	0.104	32
				S ³⁴	25.	
				S ³³	4.	
				S ³²	521.	
				<u>550.104</u>		
Cr	LZ	11.2	Cr ₂ O ₃	Cr ⁵⁴	42.	36
				Cr ⁵³	164.	
				Cr ⁵²	1,409.	
				Cr ⁵⁰	70.	
				<u>1,685.</u>		
Ni***	MB	8.7	NiCl ₂	Ni ⁶⁴	7.	40
				Ni ⁶²	21.	
				Ni ⁶¹	7.	
				Ni ⁶⁰	147.	
				Ni ⁵⁸	368.	
				<u>550.</u>		
Rb	LY	14.0	Rb ₂ CO ₃	Rb ⁸⁷	36.	8
		~17.0	Rb ₂ CO ₃ + MnO ₂	Rb ⁸⁵	97.	
					<u>133.</u>	

Table II-1. - continued

Element	Series	Process efficiency, %*	Charge material	Weight equivalent to monitored ion currents, g**	Number of collector pockets to chemistry*	
Ba***	LV	7.3	Ba metal	Ba ¹³⁸	122.	0
				Ba ¹³⁷	19.	
				Ba ¹³⁶	13.	
				Ba ¹³⁵	11.	
				Ba ¹³⁴	4.	
				Ba ¹³²	0.15	
				Ba ¹³⁰	0.21	
	<u>169.36</u>					
Pb	MA	3.7	PbCl ₂	Pb ²⁰⁸	46.	18
				Pb ²⁰⁶	22.	
				Pb ²⁰⁴	<u>1.217</u>	
	<u>69.217</u>					
Cd	LX	-	CdO	Cd ¹¹⁶	9.	0
				Cd ¹¹⁴	32.	
				Cd ¹¹³	14.	
				Cd ¹¹²	26.	
				Cd ¹¹¹	14.	
				Cd ¹¹⁰	13.	
				Cd ¹⁰⁸	0.928	
				Cd ¹⁰⁶	1.	
	<u>109.928</u>					

- * for quarter
 ** total for series to date
 *** series incomplete
 ⊕ dual collection

Table II-2. Stable isotopes separation schedule
 January - March 1964

	Building 9731		Building 9204-3		
	XAX	XBX	Track 1	Track 2	Track 3
January	Ba	W/RD	Si-S	Ca	Ni
February	Ba	W/RD	Si-S	Ca	Ni
March	Ba	W/RD	Si-S	W	Ni

Table II-3. Batches completed
October - December 1963

Element isotope	Series lot number	Weight of element, g	Assay, %	Element form
B ¹⁰ **	1(ac)95	102.0	94.7	H ₃ BO ₃
Ca ⁴² *	JW 1188(a)	21.636		CaO
Ca ⁴⁴ *	JW 1190(a)	21.601		CaO
Ca ⁴⁴ *	KZ 1330(a)	39.92		CaCO ₃
Ca ⁴⁸ *	JKL 1(a)	14.730	96.39	CaCO ₃
Cd ¹¹⁶	LT 1420(a)	10.881	97.2	CdO
Cu ⁶³ *	IW 1064(a-b)	41.72	99.62	CuO
Cu ⁶³ *	LL 1389(b)	405.83		CuO
Pb ²⁰⁴	LU 1421(a)	2.443	71.1	Pb(NO ₃) ₂
Pb ²⁰⁴	LU 1421(b)	61.419	73.3	Pb(NO ₃) ₂
Pb ²⁰⁴	LU 1421(c)	11.904	61.0	Pb(NO ₃) ₂
Pb ²⁰⁷	LU 1423(a)	762.767	92.4	Pb(NO ₃) ₂
Pb ²⁰⁷	LU 1423(b)	514.965	84.7	Pb(NO ₃) ₂
Se ⁷⁶ *	CY 398(a)	0.5438		Se
Se ⁷⁷ *	DY 515(a)	0.2885		Se
Te ¹²⁰	LK 1381(fp)	0.066	13.2	Te
Te ¹²²	LK 1382(fp)	0.458	80.4	Te
Te ¹²²	LK 1382(b)	2.740	94.4	Te
Te ¹²³	LK 1383(fp)	0.200	54.0	Te
Te ¹²⁴	LK 1384(a)	10.241	93.9	Te
Te ¹²⁴	LK 1384(fp)	0.655	83.7	Te
Te ¹²⁵	LK 1385(a)	15.088	95.0	Te
Te ¹²⁵	LK 1385(fp)	1.394	83.9	Te
Te ¹²⁶	LK 1386(a)	33.393	97.0	Te
Te ¹²⁶	LK 1386(fp)	11.076	94.0	Te
Ti ⁴⁸	KW 1316(a)-1	24.548	99.13	TiO ₂
Ti ⁴⁹ **	RS 194(c)	1.595	83.9	TiO ₂
W ¹⁸⁴ *	KQ 1284(a)	151.936	94.3	WO ₃
Yb ¹⁶⁸	LP 1401(a-1)	3.856	19.5	Yb ₂ O ₃

* returned sample

** conversion recovery

Table II-4. Special services
October - December 1963

Element	Number of samples	Type of conversion
B	3	Prepare (CH ₃) ₂ OBF ₃ for shipment
B	1	Spectrochemical analysis
B	1	Spectrochemical and mass analysis
Ba	2	Ba(NO ₃) ₂ to BaCO ₃
Ca	1	Check weight and repackage
Cd	1	CdO to metal bead

Table II-4. - continued

Element	Number of samples	Type of conversion
Cr	2	Cr ₂ O ₃ to metal
Ga	2	Ga ₂ O ₃ to metal
Hg	7	Hg(NO ₃) ₂ to Hg solution
Hg	2	Hg(NO ₃) ₂ to encapsulation
In	2	In ₂ O ₃ to In
Li	1	Grind to 325 mesh
Li	1	Spectrochemical analysis
Li	1	Repurify LiF
Mg	1	Spectrochemical and mass analysis
Mo	2	Mo to MoO ₃
Ni	1	Ni to NiO
Ni	1	NiSO ₄ to Ni
Pb	1	Pb(NO ₃) ₂ to metal button
Pb	3	Pb(NO ₃) ₂ to metal
S	1	CdS to S
Si	3	SiO ₂ to Si
Ti	2	TiO ₂ to Ti
Zn	5	ZnO to metal bead
Zn	1	ZnO to metal

III. HEAVY ELEMENT SEPARATIONS

Table III-1. Summary of Special Electromagnetic Separations

Element	Series	Process efficiency, %*	Charge material	Weight equivalent to monitored ion currents, g**	Number of collector pockets to chemistry*
U	b	16.9	UO ₂	U ²³⁸ 357.0	4
U	c	12.7	U ₃ O ₈ +C	U ²³⁸ 9.0	1

* for quarter

** for series to date

Table III-2. Operational data on completed plutonium series "a"

Innage hours	2739
Tank hours	5648
Process efficiency	13.27
Total runs	55
Average length of run	49.8 hr
Peak Q for one run	16.1 ma
Average Q for series	10.1 ma
Charge material	PuO ₂
Charge consumption rate	0.68 g/hr

Table III-3. Quantity and purity of collected isotopes, plutonium series "a"

Weight equivalent to monitored ion currents, g	Purity range, %
Pu ²⁴² 16.0052	84.22-93.76
Pu ²⁴¹ 32.2836	89.20-95.88
Pu ²⁴⁰ 126.1935	97.63-98.96
Pu ²³⁹ 71.7178	94.88-94.96

Calutron processing of plutonium was suspended in September pending recovery of feed material and these salvage operations are still in progress. Recovery and purification of the enriched isotopes has progressed to the point that 58 g of isotopic material is available for shipment (115 g Pu²⁴⁰, 28.5 g Pu²⁴¹, and 14.5 g Pu²⁴²).

A 800 mg portion of refined Pu^{241} was converted to PuF_3 used in vacuum sublimation apparatus to form Pu^{241} targets. The targets contained 44 mg Pu; 548 mg was recovered from the equipment; and 245 mg was unrecoverable from the process equipment. Development work is in progress to establish a technique for electroplating plutonium isotopes on nickel backings which will reduce process losses.

Feed material for the present U^{233} runs was prepared from two metal buttons containing 222 g of 99.6% U^{233} and 0.8 ppm U^{232} . This low-fired oxide feed was mixed with 4% graphite (by weight) prior to internally chlorinating the charge during calutron use. Two completed calutron runs yielded a total of 9 g U^{233} which is expected to have a U^{232} content of a few parts per billion. This separation is scheduled to continue until the total collection exceeds 50 g U^{233} .

Approximately 3 kg of U_3O_8 (99.98% U^{238}) was converted to UO_2 by heating a mixture containing 96% U_3O_8 and 4% C to 1100°C for 4 hr in nitrogen atmosphere. This UO_2 feed is being internally chlorinated with CCl_4 to acquire 1-2 kg of U^{238} containing 1-20 ppm of all other isotopes. Material from four receiver pockets has been recovered, but assay results have not been received.

Formation of various radioactive sources continues as needed. Electrodeposition of uranium on zincated aluminum backings yielded deposits of 2.5 mg/cm^2 on aluminum coated with $20 \text{ }\mu\text{g/cm}^2$ Zn. Other sources prepared by electrodeposition include U^{234} on Pt to provide 1700 d/sec; Am^{241} having an activity of $0.066 \text{ }\mu\text{c}$; and Am^{241} on a Pt cylinder giving $0.2 \text{ }\mu\text{c}$ activity.

Table III-4. Heavy element shipments
October - December 1963

Element	Number of shipments
Americium	13
Neptunium	6
Plutonium	11
Protactinium	1
Thorium	0
Uranium	19

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