THE
PHYSICAL REVIEW
A journal of experimental and theoretical physics established by E. L. Nichols in 1893
SECOND SERIES, VOL. 72, NO. 11 DECEMBER 1, 1947

On the Separation of Isotopes in Quantity by Electromagnetic Means

LLOYD P. SMITH, W. E. PARKINS,* AND A. T. FORRESTER†
Department of Physics, Cornell University, Ithaca, New York
(Received February 18, 1942)**

The limitations imposed by space charges on the separation of ions in the usual magnetic mass spectrograph and the possibility of trapping electrons in the ion beam are described. It is found that high voltages and intense magnetic fields are required for moderate ion currents unless these are neutralized. Calculations are given on velocity modulated or interrupted ion beams and the performance of a modulated separator is described. The theory of a radial magnetic separator is given in some detail and an experimental arrangement of such a separator proved more successful than the separator employing modulation. Some ion sources and suggested improvements are described.

INTRODUCTION

AMONG the various methods which have been used to separate small quantities of isotopes, those making use of the action of a suitable combination of static or time dependent electric and magnetic fields on ions have always been attractive. This is largely due to the fact that such methods can be used to cleanly separate almost any element as long as only very small quantities are desired.

The extension of these methods to a point where large amounts of material could be separated and still retain the advantages mentioned above has been beset with two main difficulties. The first being the production of the large ion currents necessary for the transport of a large quantity of material. So much progress has been made in ion sources, that at the present time there is no limitation set by inability to produce large numbers of ions. Consequently we shall not discuss the general construction of ion sources but confine the discussion to the particular ion sources used in the separators to be described later. The source might again in the future set a limit on the quantity of material that can be

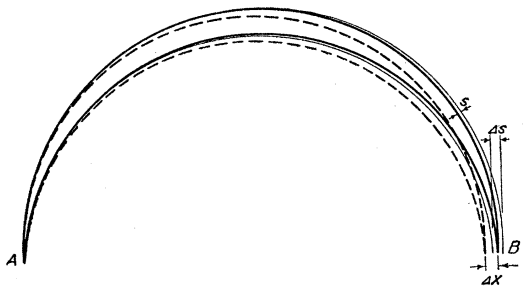

FIG. 1. Ion beams in the Dempster type mass spectrograph showing space charge divergence.

* Now located at University of Southern California, Los Angeles, California.
† Now located at RCA Laboratories, Princeton, New Jersey.
** This paper was received for publication on the date indicated but was voluntarily withheld from publication until the end of the war.

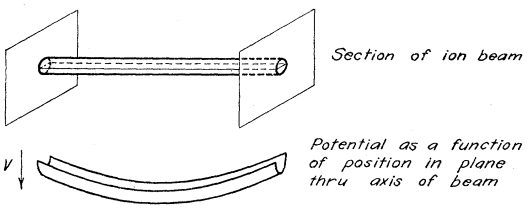

FIG. 2a. Potential distribution over a section through the axis of a beam.

separated because of velocity and space distributions of the ions, but this seems unlikely.

The second and major problem is to retain sufficient resolution in spite of the enormous space charge connected with large ion current. It may be said that as far as electromagnetic methods are concerned, their successful use for large scale separations lies in the development of a method which eliminates the large space charge forces or prevents these large forces from interfering with the applied electromagnetic forces effecting the separation.

The account to follow represents the authors' analyses and experimental investigations of promising methods conducted for the past several years which have led to a successful radial magnetic separator.

ORDINARY MAGNETIC MASS SPECTROGRAPH AND THE SPACE CHARGE PROBLEM

When a conventional mass spectrograph such as the Dempster type is considered for the purpose of separating large quantities of isotopes, one encounters a very serious limitation on the rate at which isotopes can be separated because of large space charge forces unless the space charge can be reduced in some way. An estimate of this limitation can be arrived at as follows. Consider a beam of ions of the type that would exist in a Dempster 180-degree focusing mass spectrograph, the envelope of ion paths of both isotopes being shown in Fig. 1. The beam will be assumed very long in a direction at right angles to the plane of the figure. If the change in curvature of the outermost ion path is small compared to the change in position along the center of the beam, a simple application of Gauss' theorem gives the outward force on the ions traversing the outermost paths due to space charge. This is

$$F=2\pi Ie/v, \tag{1}$$

where I is the total current in the beam per unit length perpendicular to the plane of the figure and v is the mean velocity of the ions. It is at once evident that the force on the outermost ions is independent of the thickness of the beam. Consequently the outermost ions will be subject to a constant outward force all along the beam and the trajectories of the particles on the boundary of the beam are therefore circles of radii r_1 and r_2 defined by

$$Mv^2/r_{1,2}=Hev\pm 2\pi Ie/v. \tag{2}$$

This yields for the thickness of the beam at the 180° position

$$\Delta s\equiv 2(r_2-r_1)=(8\pi e/M)(Ia^2/v^3), \tag{3}$$

where a is the unperturbed radius.

Since even without space charge the beams composed of two different isotopes focus at points very close together at B we may assume that the two beams almost coincide over the whole distance and that the outermost ions in both beams are subject to the same space charge forces. Thus both beams will have about the same thickness Δs in the neighborhood of B.

The total separation of the focal points for the two beams of isotopes at B neglecting space charge is

$$\Delta x=\frac{|M_2-M_1|}{M_1}a. \tag{4}$$

The maximum rate of deposition of cleanly separated isotopes will occur when the beam current is chosen so that the divergence Δs of each beam is equal to the separation Δx. If the spread becomes greater than the separation, i.e., $\Delta s>\Delta x$, then the area at B per unit depth of beam covered by either of the pure isotopes is Δx, and the rate of deposition of pure isotopes per unit depth of beam is proportional to $(\Delta x/\Delta s)I$. But according to Eq. (3) Δs is also proportional to I, so that nothing is gained for a current greater than that necessary to make $\Delta s=\Delta x$. From expressions (3) and (4) this gives

$$I=\frac{M_1}{8\pi e}\frac{v^3}{a}\frac{|M_2-M_1|}{M_1}.$$

Using the expression $M_1v^2/a=Hev/c$ which determines the radius of the central ion path and

the expression $v^2=2eV/M_1$, we have

$$I=\frac{1}{4\pi}\frac{|M_2-M_1|}{M_1}\frac{e}{M_1c}VH.$$

This is the limiting current. The mass $\mathfrak{M}$ deposited per unit time will be

$$\mathfrak{M}=\frac{\eta}{4\pi}\frac{|M_2-M_1|}{M_1}\frac{VH}{c}\text{ grams/sec.},$$

where η is the abundance ratio of the isotope in question. When V is in volts and H is in gausses the number of milligrams per hour that can be separated is

$$\mathfrak{M}(\text{milligrams/hr})=3.19\times10^{-8}\times\eta\frac{|A_2-A_1|}{A_1}V\text{ (volts)}\cdot H\text{ (gausses)}, \quad (5)$$

where A_1 and A_2 are the mass numbers of the isotopes. In terms of practical units and mass numbers the limiting current is

$$I(\text{ma/cm})=0.855\times10^{-6}\eta\frac{|A_2-A_1|}{A_1^2}V\cdot H. \quad (6)$$

It at once becomes evident from the above expressions that the space charge imposes a drastic limitation on the rate of separation even for very high voltages and fields. For example the rate of separation of calcium 44 from Ca 40 (a very favorable case) where $\eta=1/20$, is only about 0.12 milligrams per hour per centimeter even when $V=50{,}000$ volts and $H=15{,}000$ gausses. The total ion current in such a beam would be 1.6 ma per cm. Aside from the considerable problems attached to the use of such high voltage, it must be concluded that this method is not feasible unless the space charge can be at least in part neutralized.

In using such high voltages care would have to be exercised in designing the accelerating electrode system to prevent under or over focusing of the beam as it enters the separation region. There is also some question as to whether such high energy ions could be successfully collected at the collecting electrodes because of heating and sputtering.

Electronic Space Charge Neutralization of Beams

At first sight it would appear that effective neutralization of the large space charge in intense ion beams would be hard to realize because of the great mobility of the electrons compared to that of the ions. However an ion beam wherein the ions are not being accelerated by externally applied electric fields has the ability to trap low energy electrons so that they are confined to move along the axis of the beam. This is due to the fact that in an unneutralized ion beam the electric field is directed away from the axis of the beam and consequently the force on an electron is always toward the center of the beam. The ion beam forms a space charge trough, so to speak, and low energy electrons once in the beam cannot get far away from the axis. They can however move along the axis of the beam but even near the ends of the beam the electrode potentials can be arranged so as to prevent low energy electrons once in the beam from getting out. The potential distribution in an ion beam is shown in Fig. 2a. If electrons can be successfully trapped then a large number can accumulate and effectively reduce the space charge.

When a magnetic field is present as in the Dempster type mass spectrograph, the electrons can be even more effective and migrate throughout the beam even though a strong magnetic field is present. Since there is a force on the electron toward the center of the beam, an electron initially at rest in the beam will execute very small cycloid-like paths as shown in Fig. 2b. If at any time it should lose kinetic energy by suffering a collision, it will move nearer the center of the beam. If near the end of the beam there is a small retarding force the electron will move across the beam to the other side and return as shown. In this way electrons can eventually migrate to any part of the beam.

The mean velocity of the electron along the beam would be of the order $v_e=(E/H)c$, where E is the electric field at right angles to the beam In an unneutralized beam E could be of the order

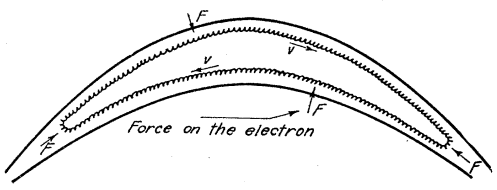

FIG. 2b. Possible electron path through the ion beam in an ordinary mass spectrograph indicating the trapping of slow electrons.

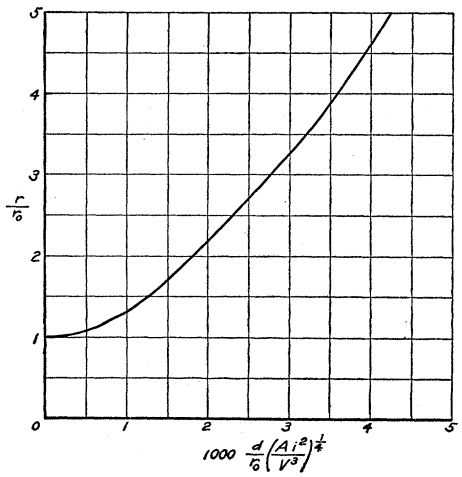

FIG. 3. Natural divergence of a beam of circular cross section in which only radial forces are assumed. r_0 is the original radius; r the final radius; d, distance traveled; i, total current; V, initial voltage; and A, the atomic weight of the particles. Practical units used throughout.

of a thousand volts per cm and the migration would take place quite rapidly. However, if we suppose the ion density is not high, or the beam has been partially neutralized already, and take E to be even 10 volts per cm, then in a field of 2000 gausses,

$$v_e \approx 5\times 10^5 \text{ cm/sec.}$$

An ion of mass number 200 would have a velocity of the order

$$v_i \approx 10^5\ V \text{ (volts)},$$

where V is the energy of the ions in electron volts. This might be of the order 3600 volts for instance. Then

$$v_i \approx 6\times 10^6 \text{ cm/sec.}$$

which is some 10 times the migration velocity of the electrons. Consequently these electrons could be very effective in neutralizing space charge. Such electrons might be introduced into the beam by ionization of gas atoms, or by appropriately placed electron emitters.

MODULATION METHODS AND THE SPACE CHARGE PROBLEM

Methods which make use of a time variation of electric fields for effecting isotope separation are also severely influenced by space charge. In these methods usually an attempt is made either to velocity modulate a uniform beam of ions or to interrupt the beam periodically and then make use of a drift space to obtain separation. In such cases the space charge gives rise to two difficulties. One is the rapid space charge divergence of the beam and the other has to do with the shielding effect of ions surrounding the modulating electrodes.

The divergence of a beam in the form of a sheet of infinite width is given essentially by an expression already used. If d is the distance along the beam measured from a point where the ions were traveling in parallel paths, then the divergence is given by

$$\frac{z}{z_0} = 1 + 2.04\times 10^6 \frac{d^2 I}{z_0}\frac{A}{V^3}, \tag{7}$$

where z_0 is the original thickness of the beam, I is the current per unit length in the infinite direction in amperes, V the voltage of the beam, and A is the atomic weight of the ion.

When the beam has cylindrical cross section the expression is a more complicated function of the distance d along the beam. This expression has been derived by several authors[1] and may be written in the form

$$d = 1.75\times 10^{-3}\, r_0 \left(\frac{V^3}{AI^2}\right)^{\frac{1}{4}} \int_0^{\log r/r_0} \exp\,[y^2]dy, \tag{8}$$

where I is the beam current, V energy of ions in electron volts, r_0 the initial radius, r the radius at distance d, and A is the atomic weight of the ion. For convenience a plot of this function is given in Fig. 3. As an example of the order of magnitude of the divergence to be expected in useful beams, it is readily found from Fig. 3 that a 10-ma, 10,000-volt beam of ions of mass 40 will diverge from an initial radius of 1 cm to a radius of 3 cm in a distance of 5.6 cm.

If parallelism of ion paths is important, it is evident that unless high initial voltages are used practical currents for separation are impossible. If high voltages are used one encounters most of the problems previously discussed in connection with high voltage beams. A further complication is introduced in the modulation of high voltage beams because this requires that the frequency and amplitude of the

[1] See for instance: L. P. Smith and P. L. Hartman, J. App. Phys. **11**, 220 (1940).

modulation field must be made high. In any practical arrangement this turns out to be a source of difficulty and especially so if any modulation voltage other than a sinusoidal one is desired. Another difficulty with modulation schemes is the necessity for sharp potential gradients in certain regions. This may be difficult in a region of high space charge, even when grids are used.

The use of a magnetic field in the direction of the beam to prevent divergence is not very effective as is shown below. Calculations can be made for the maximum radius attained by a particle on the outside of a beam of circular cross section. Such a particle moves periodically between a maximum and minimum radius as it progresses in the axial direction. Except for this axial motion, conditions are not unlike those in a magnetron. Hull[2] has shown that, for a charged particle moving in a homogeneous magnetic field and a symmetrical radial electric field F (function of r only, the magnetic field being in the z direction), where initial conditions at $r=r_0$ are taken to be

$$\mu_0=(dr/dt)_0, \quad v_0=r_0(d\theta/dt)_0, \quad w_0=(dz/dt)_0, \tag{9}$$

the radial velocity squared may be written

$$\left(\frac{dr}{dt}\right)^2=\frac{2eV}{m}-H^2\left(\frac{e}{2m}\right)^2r^2\left(1-\frac{r_0^2}{r^2}\right)^2 -H\frac{e}{m}r_0v_0\left(1-\frac{r_0^2}{r^2}\right)+v_0^2\left(1-\frac{r_0^2}{r^2}\right)+u_0^2, \tag{10}$$

where

$$V_r=\int_{r_0}^{r}F dr. \tag{11}$$

Putting $dr/dt=0$, Hull obtained an implicit expression for the maximum radius r_m which the particle can reach, i.e.,

$$V_{r_m}=H^2\frac{e}{8m}r_m^2\left(1-\frac{r_0^2}{r_m^2}\right)^2 +\left(\frac{Hr_0v_0}{2}-\frac{v_0^2}{2e/m}\right)\left(1-\frac{r_0^2}{r_m^2}\right)-\frac{u_0^2}{2e/m}. \tag{12}$$

In order to find the maximum radius reached by the beam confined by an axial magnetic field, we shall determine that of an ion starting on the edge of the beam at r_0. For this maximum radius the ion can be assumed to remain on the outside of the beam as it diverges outward. From Gauss' theorem the effective radial electric field expressed in terms of the beam current I, is

$$F=\frac{2Ic^2}{rw_0}.$$

Making use of the expression (11) we find

$$V_{r_m}=\frac{2Ic^2}{w_0}\log\frac{r_m}{r_0}. \tag{13}$$

Substituting (13) in Eq. (12) and letting $v_0=0$, the maximum radius can be determined from the equation

$$\frac{2Ic^2}{w_0}\log\frac{r_m}{r_0}=\frac{H^2er_0^2}{8m}\left(\frac{r_m^2}{r_0^2}-2+\frac{r_0^2}{r_m^2}\right)-\frac{m}{2e}u_0^2. \tag{14}$$

On setting $\rho=\log(r_m/r_0)$, this equation becomes

$$\frac{2Ic^2}{w_0}\rho=\frac{H^2er_0^2}{2m}\sinh^2\rho-\frac{m}{2e}u_0^2. \tag{15}$$

Changing (15) from e.m.u. to practical units gives

$$2.72\times10^{10}\frac{A^{\frac{1}{2}}I}{(V_z)^{\frac{1}{2}}H^2r_0^2}\rho+2.09\times10^4\frac{AV_R}{H^2r_0^2}=\sinh^2\rho. \tag{16}$$

A is the atomic weight of the singly charged ions,

$$V_z=\left|\frac{mw_0^2}{2e}\right|, \quad \text{and} \quad V_R=\left|\frac{mu_0^2}{2e}\right|.$$

Solutions for r_m are most easily obtained by first finding a graphical solution for ρ. To illustrate the magnitude of the maximum radius for certain conditions, consider a particle with only z velocity on the edge of a 1-cm diameter, 10-ma, 10,000-volt beam of mass 40 ions. If the magnetic field is 5000 gauss, this radius is 18.6 cm. It is also important to know how rapidly the beam

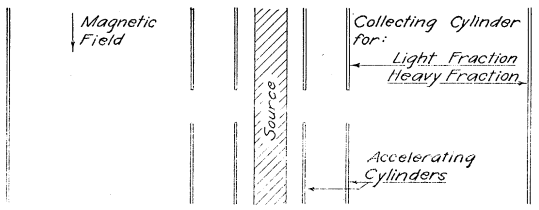

FIG. 4. Schematic diagram for radial magnetic separator.

[2] A. W. Hull, Phys. Rev. **18**, 31 (1921).

diverges. To do this would require a determination of the time required for a particle to reach the maximum radius. For our purposes this may be avoided in the following manner. It may be said that until the ion has reached a distance of about one-half the maximum radius the magnetic force in the r direction is small compared to the electric force. It is plausible to assume therefore that until the ion has reached a radius of about $\frac{1}{2}r_m$, the divergence would be fairly close to that produced by the space charge alone as given in Fig. 3. Consequently the magnetic field cannot be regarded as an effective preventive for space charge divergence of an ion beam.

An axial magnetic field would only prove useful if slow electrons were allowed to be present in the beam. These electrons could be confined to any cross-section beam and furnish the necessary negative space charge to prevent divergence of the ions. This principle has been used by Finkelstein[3] in an intense ion source.

Possible Use of Divergence

In view of the space charge troubles in the operation of modulation devices, an interesting modification suggests itself. This would involve making use of space charge divergence in the actual separation. The natural divergence of a group of charged particles will carry the lighter particles to greater radial distances than it does heavy particles in the same time. Collectors arranged in the proper positions could collect only particles of one mass. An alternative method of operation would be to apply certain potentials to a set of collectors and make use of the fact that the lighter particles of successive bunches also acquire greater energies as the bunch diverges. As far as is known, no separation devices have been designed on these principles, but it presents an interesting possibility. The chief difficulty with such a method would be in producing periodic bunches of space charge before the divergence took place.

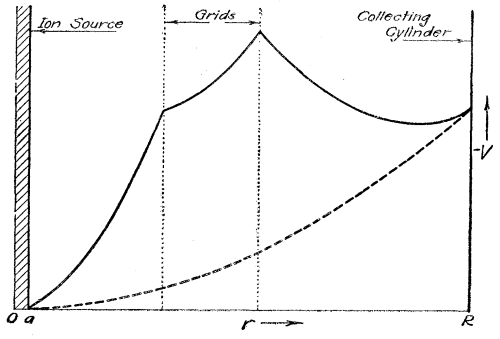

FIG. 5. Potential distribution showing critical condition at the collecting cylinder.

[3] A. T. Finkelstein, Rev. Sci. Inst. **11**, 94 (1940).

THE RADIAL MAGNETIC SEPARATOR AND THE SPACE CHARGE PROBLEM

Attempts to devise an instrument whose resolution would be independent of space charge, at least to the first order led to the radial magnetic separator. As shown in Fig. 4, the ions are produced at the center of the apparatus and are accelerated radially by one or more cylindrical grids or slits. Under the action of an axial magnetic field the particles describe orbits which depend on their masses in such a manner as to permit the collection of the heaviest ions on a cylindrical electrode, of diameter large compared to the source, while the remaining light fraction is turned back towards the center and can be collected by electrodes of suitable geometry.

The advantageous feature of this method lies in the fact that the potential required to turn back a particle after it has traveled out to some radius depends only on the radius and, as can be seen from Eq. (12), not at all on the variation in potential over the region through which it has traveled. Therefore, except insofar as the space charge produces fields in the axial direction, it has no effect, until the raising of the potential due to space charge is sufficient to stop some heavy ions before they reach the outer cylinder. It is to be emphasized that although this provides a limitation on the current, as calculated below, it does not alter the fact that only the heaviest ions can reach the outer cylinder and that the remaining fraction, enriched in light ions, can be collected as it moves back towards the center.

If the length of the apparatus is not large compared with its diameter, then space charge forces will produce fields in the axial direction. Since this interferes with the resolution only insofar as it produces a spread in radial velocities, its effect is secondary.

A feature not to be overlooked in this apparatus is that a single ion source can be used to feed an extended apparatus.

An estimate of the current limitation imposed by space charge may be arrived at by the following analysis. For a given set of conditions (positions of the electrodes, their potentials with respect to the ion source, and the strength of the magnetic field) the current to the outer cylinder has a limiting value determined by the condition that the magnitude of the potential at all points between the collecting cylinder and the source must be greater than that of the potential given by Eq. (12), i.e.,

$$V_0=-\frac{1}{8c^2}\frac{e}{M}H^2r^2\left(1-\frac{a^2}{r^2}\right)^2.$$

H is in gauss and all other quantities in e.s.c.g.s. units. Here a is the distance from the center at which the charged particles start. It is assumed that all the particles start at the same distance from the center with zero velocity. V_0 is the potential taken with respect to their starting position. Since we are interested only in distances for which

$$a/r\ll 1,$$

we may use

$$V_0=-\frac{1}{8c^2}\frac{e}{M}H^2r^2. \tag{17}$$

The nature of the problem is made clear by reference to Fig. 5, which shows a source of small diameter surrounded by grids to supply sufficient potential to remove ions and send them to the outer cylinder. The problem of removing sufficient ions from the source out to the 1st grid, in the case of an arc type source, which is a practical necessity if large ion currents are desired, is automatically taken care of. The space charge in the arc must increase sufficiently to force most of the ions formed radially outwards. We shall be concerned only with the region between the outermost grid and the collecting cylinder, where we wish to find what the distribution is for the critical conditions. From this we can find what is the minimum potential which must be placed on a grid at a fixed distance from the center to obtain a given current to the outer cylinder.

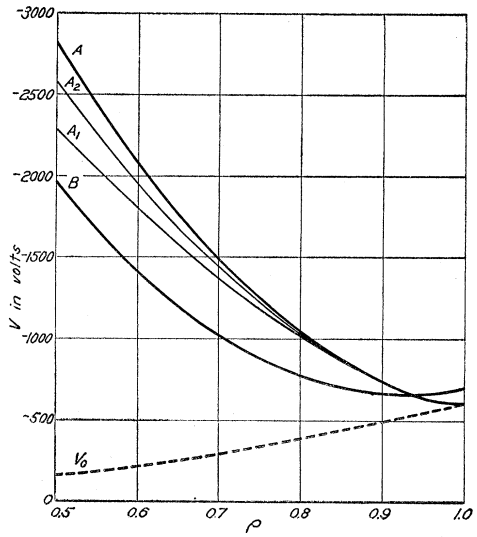

FIG. 6. Potential distributions for critical conditions as a function of the fractional distance from the center to the collecting cylinder for Li^7 ions for: 3.63 ma per cm; 1350 gauss; 14 cm radius. A shows the distribution when the ions reach the outer cylinder with zero radial velocity; B when the potential is adjusted to just stop the Li^6 ions. Both were obtained numerically. A_1 is the approximation obtained to A by taking the first term and A_2 by taking all of the computed terms of the series. The effect of Li^6 ions on the potential has been neglected.

If the ions strike the cylinder with zero radial velocity, i.e.,

$$V(R)=V_0(R), \tag{18}$$

then the limiting current condition is

$$V'(R)=V_0'(R). \tag{19}$$

If symmetry eliminates the z and φ coordinates from Poisson's equation, we have

$$\frac{1}{r}\frac{d}{dr}r\frac{dV}{dr}=-4\pi\rho. \tag{20}$$

Since

$$I=2\pi r\rho v_r$$

is the radial current per unit length, we obtain

$$\frac{d}{dr}r\frac{dV}{dr}=-\frac{2I}{v_r}.$$

The radial motion is governed by the Brillouin potential[4] P, i.e.,

$$M\frac{d^2r}{dt^2}=-e(\partial P/\partial r),$$

[4] Leon Brillouin, Phys. Rev. 60, 385 (1941).

where

$$P = V - V_0.$$

In terms of this variable our differential equation is

$$\frac{d}{dr}r\frac{d}{dr}(P+V_0) = \frac{-2I}{(-2eP/M)^{\frac{1}{2}}},$$

which becomes

$$\frac{d}{dr}r\frac{dP}{dr} - \frac{1}{2c^2}\frac{e}{M}H^2 r = -I\left(\frac{2M}{eP}\right)^{\frac{1}{2}}. \tag{21}$$

From (18) and (19) we get the boundary conditions

$$P(R) = 0; \qquad P'(R) = 0. \tag{22}$$

The differential equation can be simplified by the use of the dimensionless variables

$$\rho = r/R, \quad \Phi = \alpha P, \quad \alpha = \left(\frac{e/M}{2I^2R^2}\right)^{\frac{1}{3}}.$$

Equation (21) becomes

$$(-\Phi)^{\frac{1}{2}}\left\{\frac{d}{d\rho}\rho\frac{d\Phi}{d\rho} - K\rho\right\} = -1, \tag{23}$$

where

$$K = \left(\frac{(e/m)^2R^2}{4I}\right)^{\frac{2}{3}}\frac{H^2}{c^2}.$$

The boundary conditions for Φ are

$$\Phi(\rho) = 0; \qquad \Phi'(\rho) = 0 \qquad \text{at } \rho = 1. \tag{24}$$

If we write (23) in the form

$$(-\Phi)^{\frac{1}{2}}\{\rho\Phi'' + \Phi' - K\rho\} = -1,$$

it becomes apparent that $\Phi'' \to \infty$ as $\rho \to 1$. Therefore as $\rho \to 1$, the solution of this equation approaches the solution of

$$\Phi_0''(-\Phi_0)^{\frac{1}{2}} = -1; \quad \Phi_0(1) = 0; \quad \Phi_0'(1) = 0.$$

This is readily seen to be satisfied by

$$\Phi_0 = -(9/4)^{2/3}(1-\rho)^{4/3}. \tag{25}$$

Φ_0 is close to the exact solution of (23) where

$$\rho \approx 1$$

and

$$|\Phi_0' - K| \ll \Phi_0''.$$

For a given value of K the approximate solution can be used as a starting point for a numerical solution of (23), and from Φ, the potential

$$V = P + V_0 = \frac{\Phi}{\alpha} + V_0$$

can easily be found. This is a tedious process and each numerical integration is valid for only one choice of K.

In order to find a series solution substitute

$$u^2 = -2^{\frac{2}{3}}\Phi = -2^{\frac{2}{3}}\alpha P$$

and obtain the differential equation

$$u\frac{d}{d\rho}\rho u\frac{du}{d\rho} = 1 - K'\rho u, \tag{26}$$

where

$$K' = K/2^{\frac{1}{3}}.$$

From Φ_0 we get the solution to this differential equation valid for ρ close to one,

$$u_0 = (9/2)^{\frac{1}{3}}(1-\rho)^{\frac{2}{3}}. \tag{27}$$

This suggests a solution of the form

$$u = \sum_{k=2}^{\infty} c_k(1-\rho)^{k/3}. \tag{28}$$

In Eq. (26) let

$$x = (1-\rho)^{\frac{1}{3}} \tag{29}$$

and try a solution of the form

$$u = c_2x^2 + c_3x^3 + c_4x^4 + \cdots. \tag{30}$$

Values of several of the constants are:

$$\begin{aligned}
c_2 &= (9/2)^{\frac{1}{3}},\\
c_3 &= 0,\\
c_4 &= -\frac{K}{20}(9)^{\frac{2}{3}},\\
c_5 &= \frac{4}{15}\left(\frac{9}{2}\right)^{\frac{1}{3}},\\
c_6 &= -\frac{27}{14}\frac{K^2}{100}\frac{1}{2^{\frac{2}{3}}},\\
c_7 &= -\frac{28}{405}(9)^{\frac{2}{3}}K,\\
c_8 &= \left(\frac{9}{2}\right)^{\frac{1}{3}}\left\{\frac{67}{405} - \frac{827}{1008}\frac{K^3}{1000}\right\}.
\end{aligned} \tag{31}$$

The series converges slowly and is therefore not very satisfactory. For a specific case, chosen

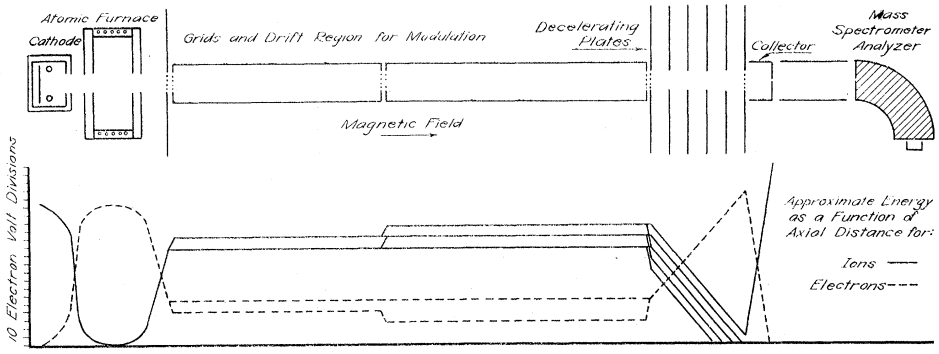

FIG. 7. Schematic diagram of isotope separator making use of velocity modulation. Approximate applied potentials may be obtained from the energy diagram.

because it can be compared with available experimental data, the value has been obtained and plotted in Fig. 6. There the potential obtained from u_0 (curve A_1) and that obtained using all of the terms of the series for which the coefficients have been calculated (curve A_2) are compared with the exact solution (curve A) obtained numerically.

The requirement that particles strike the outer cylinder with zero radial velocity is too stringent, for in practice they would be collected with some radial velocity. Let us suppose that the potential on the outer cylinder is adjusted to the correct value to just stop the lighter isotope, in the case for which the calculations have been made, the Li^6 ions.

$$V(R)=\frac{-1}{8c^2}\frac{e}{M_6}H^2r^2, \tag{32}$$

where M_6 represents the mass of the Li^6 ions. Take for the other boundary condition, as before,

$$V'(R)=V_0'(R),$$

although in this case this condition is too severe.

For this case it might be expected that an ordinary power series could be used, but the solution so obtained converges so slowly as to be altogether impractical. The result of numerical integration can be seen in curve B in Fig. 6.

For the range of ρ covered in Fig. 6, A_1 is a sufficiently good approximation to the exact solution, so that we may use the analytic expression for A_1 to find the manner in which the potential varies with I, e/M, H, and R. It is also reasonable to suppose that curve B varies in approximately the same manner as curve A.

The expression for A, is, from Eq. (25)

$$V=V_0+\Phi_0/\alpha$$
$$=-\left\{\frac{1}{8c^2}\frac{e}{M}H^2r^2+\frac{81}{8}\frac{I^2R^{\frac{1}{3}}}{e/M}(1-\rho)^{4/3}\right\}.$$

Except for ρ close to 1, V_0 may be neglected in comparison with Φ_0/α. We then obtain the proportionality equation

$$V\sim I^{\frac{2}{3}}R^{\frac{2}{3}}M^{\frac{1}{3}} \tag{33}$$

for a fixed value of ρ. It will be noticed that to this order of approximation V is independent of the magnetic field.

EXPERIMENTS ON A MODULATION SEPARATOR

After consideration of the advantages and disadvantages of the various separation methods mentioned in the foregoing analysis two schemes appeared sufficiently promising to be investigated experimentally. The first of these is a velocity modulation method making use of electron space charge neutralization.

The apparatus is schematically shown in Fig. 7. All of it except for the mass spectrograph analyzer is located inside a series of coaxial coils which provides a magnetic field of a few hundred gausses. The ion source is modeled after that described by Finkelstein.[3] The system is designed so that the electrons which produce the ions also furnish the space charge neutralization. They are accelerated from the cathode, pass

through the atomic beam, and on through the system to the ion collector where they are repelled and sent back; whereafter, if they have not lost too much energy, they are available for producing still more ions. If these electrons have lost any energy on their trip to the ion collector and back, they cannot reach the cathode again, but will be forced to travel back and forth through the system furnishing negative space charge and producing ions until they are lost out of the beam. The approximate energy of the electrons as a function of distance is shown in Fig. 7 also. Because of low electronic mass each trip through the modulation region takes place so quickly that only in rare cases will the electron's energy be modified by its passage through this region. The magnetic field serves to hold the electrons in a fairly well defined beam. The potential trough formed by the electrons in turn serves to keep the ions from diverging.

Ions are produced by electron collision in the atomic jet and are sent through the modulation region, but for the ions the energy with which they emerge depends on their mass. The frequency and amplitude of the modulation voltage as well as the initial ion energy and drift region lengths are adjusted to allow only one mass ion to emerge with increased energy. Next the ions pass through a set of electrodes having progressively higher potentials. This slows the ions and finally causes all of them to stop except those which have received the increased energy in the modulation region, these being only ions of the desired isotope. The ions stopped before the last

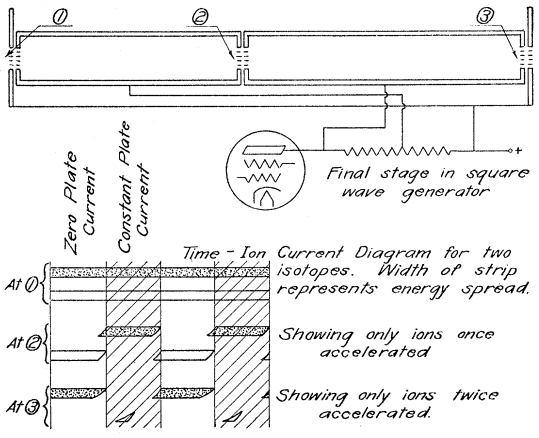

FIG. 8. Modulator and its principle of operation.

grid for the most part diverge and are collected on the nearby electrodes, some may return down the system, but this number will be small. To aid in the adjustment for obtaining the best separation conditions, a mass spectrograph is used to analyze the ions passing through a small opening in the collector.

The potential arrangement requires the arc drop (cathode to atomic jet) be greater than the initial voltage given the ions as they pass into the modulator. This requires low voltages as long as the ion beam can be held together. With low ion energies modulation with a sinusoidal voltage cannot be very effective, but at the same time other wave forms are not difficult to obtain for low amplitudes. A square wave is most desirable here, making possible the pure separation of almost half of the ions of the desired type leaving the source. Such a square wave is used, produced by a generator of the type described by Parkins and Smith.[5] Because of slight ion energy spread due to the source, completely pure separation of the desired isotope cannot be obtained here in less than three modulations. Three pairs of grids are used, the connections made to the load resistor in the square wave generator as indicated in Fig. 8. This figure also diagrammatically shows the principle of giving increased energy to ions of only one isotope.

The cathode is a dispenser type and a modification of that described by Hull.[6] One loop of molybdenum stocking containing the $BaO\text{-}Al_2O_3$ eutectic serving as the heater and dispenser is centered in front of a flat molybdenum emitting surface. The hole in the front of the cathode shield determines the beam diameter and was made $\frac{1}{4}$ in. smaller than the molybdenum stocking loop so that no bombardment of the stocking could take place.

The furnace is a double-walled cylindrical unit made from molybdenum sheet. A tungsten coil with insulating refractory beads is placed between the walls and serves to vaporize the material to be separated. Calcium was used in the experiments and was simply put in the metallic state on the floor of the furnace.

Grids are of $\frac{1}{16}$-in. wide tantalum ribbon, all

[5] W. E. Parkins and Lloyd P. Smith, Phys. Rev. **57**, 108 (1940).
[6] A. W. Hull, Phys. Rev. **56**, 86 (1939).

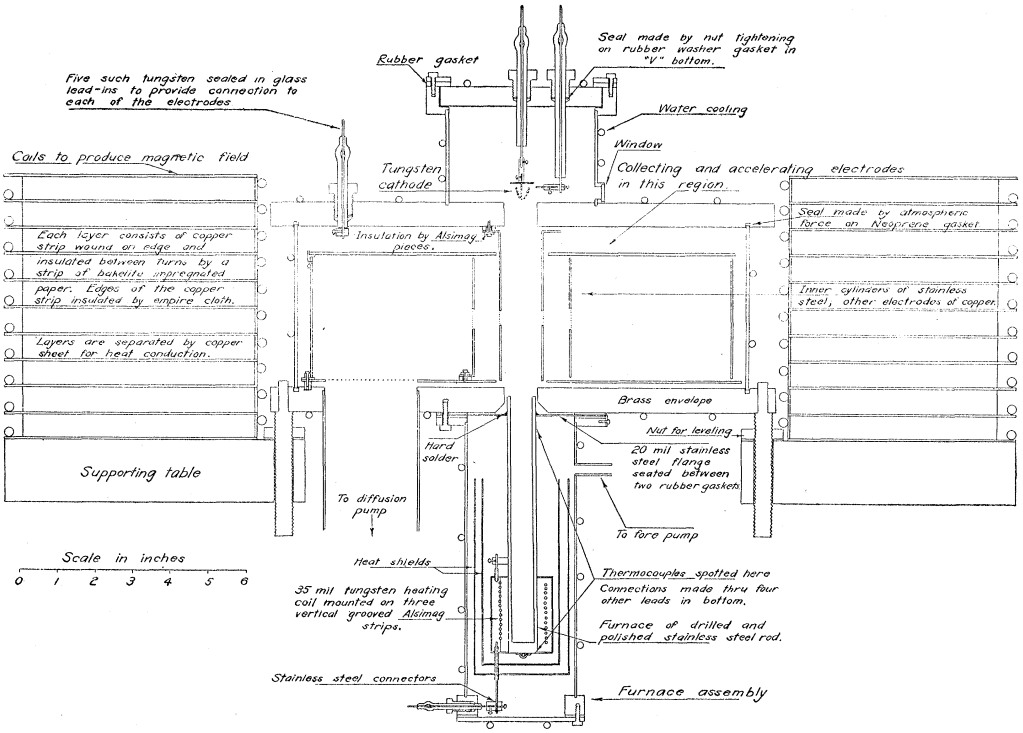

FIG. 9. Apparatus used in the investigation of the radial magnetic separator showing typical electrode arrangement.

lined up in winding and presenting only their narrow edges to the beam. The drift region shields are of $\frac{1}{2}$-in. diameter stainless steel tubing adjustable in length. The plate electrodes are of 20-mil non-magnetic stainless steel.

The entire unit, only 18 in. long, is mounted on three glass rods fixed in a brass base plate. Tungsten sealed in glass leads are brought in through the base plate making use of rubber gaskets as shown in Fig. 9. To save space one glass tube carries eight of the low current leads. The base plate with all electrodes is assembled and then placed inside the system, the seal being made at the base plate rim with a rubber gasket. The mass spectrograph analyzer is a permanent part of the system.

In the operation of this modulation device it was not difficult to produce an arc and obtain large ion currents at the input end of the modulator. However, only extremely small ion currents were ever measured at the collector. The difficulty with the scheme lies in obtaining effective electron neutralization of the ion beam, and this is the feature upon which the attainment of large transport in low energy ion beams depends. Reasons for ineffective electron neutralization are:

1. Too many variations in electron energy are present.
2. The arc drop is so difficult to adjust that an independently controlled initial electron energy is impossible.
3. At the points where the electrons are turned back too many of them are lost. This takes place near the collector because the increased negative space charge causes the attainment of larger radii as predicted by Eq. (15). Near the cathode loss of electrons takes place by scattering due to the fact the electron collision cross section is so high for low energy electrons.

EXPERIMENTS ON THE RADIAL MAGNETIC SEPARATOR

From Eq. (12) it can be seen that a pure separation of two isotopes can be made in the radial magnetic separator only if the contributing ions are formed at positions having a limited variation in radii and potentials. For a pure separation the voltage spread allowable in the source is equal to the difference in critical voltages for the ions of the two isotopes if all particles are

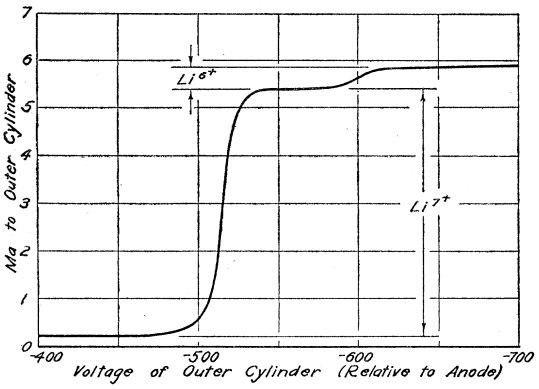

FIG. 10. Current-voltage characteristic of the outer cylinder of the radial separator obtained with the electrode arrangement in Fig. 6.

assumed to start from the same initial radius with zero energy. Energies of ions upon formation may always be neglected. Should all ions be formed in a region of constant potential, the allowable spread in starting radii may be found from:

$$\frac{r_m^2+2r_{01}^2+(r_{01}^2/r_m^2)}{r_m^2+2r_{02}^2+(r_{02}^2/r_m^2)}=\frac{M_1}{M_2},$$

where M_1 and M_2 are the masses of the two types of ions, r_{01} and r_{02} are their initial radii, and r_m is the radius of the outer cylinder. This equation was obtained by equating critical voltages. Choosing an r_0 for ions of one mass, for instance r_{01}, the two solutions for r_{02} will define the range of allowable initial radii of ions of the other mass for complete separation. If a source is used in which the initial radius and potential both vary, the calculation of conditions for complete separation will have to include both effects.

There are other factors which may cause mixing of the isotopes. These may be listed:

1. Excessive deviations of the applied electric field from a purely radial field,
2. Non-uniformities in the magnetic field,
3. Asymmetry in space charge fields,
4. Space charge potential variation in the z direction in the separation region,
5. Space charge oscillations which succeed in varying the energies of the particles.

The first factor may always be overcome by improving the electrode geometry and choice of potentials, using grids if necessary. The second presents even less of a problem; a sufficiently homogeneous magnetic field is easy to obtain. The last three factors concern space charge effects and all have been found to be present. It is difficult to estimate the extent to which these effects will influence the resolution.

For the experimental investigation of the radial magnetic separator lithium was used. The vertical magnetic field made possible an ion source of definite radius by providing a means for confining an electron beam. A cathode was placed on the axis of the system facing a furnace from which lithium was vaporized. With the furnace as the anode a low voltage arc could be struck, from which considerable ion current could be drawn off in the radial direction.

Dispenser and oxide coated cathodes were tried, but not found superior to the simpler tungsten type. Because of the chemical activity of molten lithium and the necessity for the use of non-magnetic materials throughout, the furnace was made of stainless steel. Means of heating the furnace in air were not very successful, but enclosing the furnace in a separate vacuum chamber and heating by a tungsten coil proved to be satisfactory. To conserve lithium and prevent excessive scattering, a collimated atomic beam was obtained and the material removed in collimation returned to the reservoir. This was accomplished by use of a long vertical furnace heated at the bottom. The temperature at the top of the furnace was not far above the melting point of lithium so that much of the vapor impinging on the walls was condensed and returned by gravitational flow. The furnace could not be narrowed at any point or the molten lithium would cling and close the opening. Also, it was found the inside walls had to be smooth to assure satisfactory refluxing action. A platinum coating applied to the inside of the furnace was helpful in this respect.

To obtain samples of the lithium isotopes the collecting surfaces in the separation chamber must be sufficiently cool to condense the material. The early experiments were done with the first cylinder (numbered from the inside) water cooled to prevent heat from the arc and collected current from raising the electrode temperatures excessively. This, however, did not allow the flexibility of potentials necessary in these first measurements. All electrodes (three cylinders and

two end plates) were then insulated from one another and from the metal envelope so that potentials could be controlled and currents measured individually. The first two cylinders were used in a variety of forms, experimenting with grids, slit sizes, and cylinder radii. A typical arrangement for the entire system is shown in Fig. 9.

To determine the success of the separation and optimum operating conditions, current to either the second or third cylinder was plotted against voltage on the third cylinder arc and magnetic field conditions remaining constant. When this voltage is too positive, ions will not arrive at the third cylinder, but return for the most part to the second. As the third cylinder becomes sufficiently negative, ions of the heavy isotope arrive, increasing the current there and decreasing it at the second cylinder; at a still more negative voltage, ions of the light isotope cause a similar variation in currents to these cylinders. As a result the current to either cylinder as a function of the voltages applied to the third cylinder has plateaus to be identified with the ions of each isotope present. The fact that the magnitude of the differences in current between plateaus are proportional to the relative abundance of the isotopes in question constitutes a check on their identity. In Fig. 10 a current-voltage characteristic taken for the third cylinder is reproduced.

Not only do these characteristics furnish a measure of the quality of the separation, but they are useful in forming a better understanding of the action of the separator. Characteristics for the other electrodes are helpful in this connection. To save the tedious point by point plotting, an oscilloscope may be used to trace the curves. For this scheme a 60-cycle voltage is connected in series with the d.c. voltage applied to the third cylinder, and is also connected across the horizontal plates of the oscilloscope. A resistance carrying the current to the electrode in question is connected across the vertical plates. During long runs while separation is in progress such an oscilloscope would be invaluable in checking operating conditions, the a.c. need only interrupt the separation process for a few seconds.

One of the greatest difficulties proved to be suppression of oscillations which were spontaneously generated and found on all electrodes. Various connections of capacitances and inductances were never uniformly successful in reducing their intensity. It was finally found they arose in the arc region and could be removed only by proper control of the space charge conditions there. Stopping all oscillations also removed peculiar space charge potential distribution in the arc region which was shown to be present by measurement of current to the various electrodes. This proper control was brought about by keeping the arc current fairly small (below $\frac{1}{2}$ ampere) and putting a negative voltage (usually 500 or 600) on the first cylinder. This potential caused considerable current to be drawn from the arc, more the larger the arc current. Making the first cylinder sufficiently negative would, of course, finally extinguish the arc. At the same time the accelerating voltage on the first cylinder was desirable because it increased the current passing into the separation region. Actual ion currents removed from the arc to the first cylinder and separation regions were often as great as one third of the arc current. The fact that these measured currents were ion currents was assured by the effect of the first

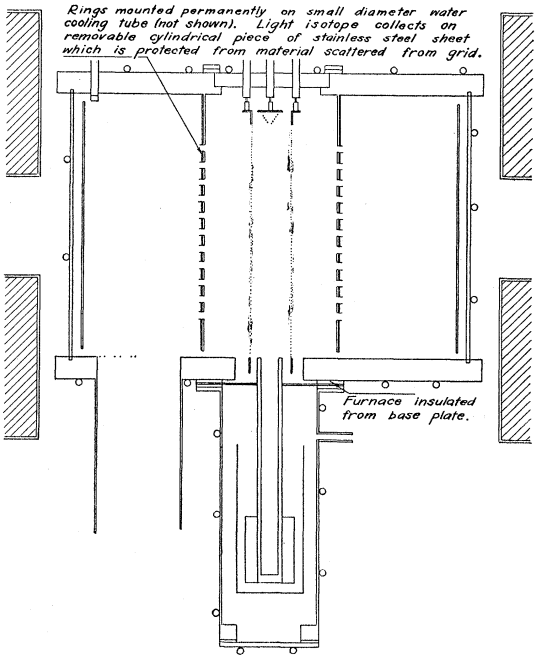

FIG. 11. Modification of the apparatus recommended for the quantity collection of pure Li^6.

cylinder potential on the saturation current to the third cylinder. For a second check it was noted that the ratio of current to the first cylinder to the current through the first slit was always approximately equal to the ratio of the effective length of the first cylinder to the length of the slit.

Through a $\frac{3}{8}$-in. slit in the first cylinder, 8-ma saturation currents to the third cylinder were shown to be separated by a characteristic not unlike that in Fig. 10. Larger currents could be brought into the separation region but not without altering the arc sufficiently to bring about the undesirable space charge conditions, and with them apparent incomplete separation of the isotopes. Through a $3\frac{1}{4}$-in. slit (only grids used for the first and second cylinders) 30 ma were shown by the third cylinder characteristic to be well separated. In this latter case currents to the end plates were large. Both measurements quoted were made with the end plates and second cylinder at the same potential as the third cylinder.

Experiments were also made with a source consisting of a nickel cylinder coated with spodumene and mounted concentric with the system axis at the slit. When the cylinder was heated, as much as 1 ma was obtained through a $\frac{3}{8}$-in. slit, giving good separation. From the spodumene some sodium and potassium ions were observed. Other ions were observed with the arc source also, in particular Li_2^+.

Figure 11 shows a proposed apparatus for the separation of Li^6. Provision is made for the use of current from an extended region of arc. Bombardment of the first grid will be sufficient to prevent lithium from condensing and clogging the openings, and at the second cylinder, which is cooled, the use of grids is avoided. The top and bottom of the system take the place of the end plates. These are at the potential of the second cylinder, and it is expected this whole unit will be operated at the third cylinder potential. The third cylinder is insulated to make possible the measurement of current-voltage characteristics.

Calculated *versus* Actual Space Charge Limitations

The results of the space charge calculations made in the previous sections and shown in Fig. 6 correspond to actual ion currents of lithium for which clean cut separation of Li^6 and Li^7 was achieved. According to the calculations the potential at a point half-way between the ion source and outer collecting cylinder should be of the order of 2000 volts below the source potential in order that a current of 3.63 ma per cm be collected at the outer cylinder. However such currents were actually measured when the potential there was less than 600 volts below the source potential. While it is true that the actual apparatus is limited in length, the influence of the end plates on the potential at the center cannot be appreciable since the distance between end plates is comparable to the distance between the source and the outer cylinder. Also since there was very little ion current to the end plate compared to that to the outer cylinder it must be concluded that the positive space charge in the separation region is not as great as it would have to be if only the positive ions making up the current to the outer wall were present there. Consequently it must be concluded that the space charge is neutralized in some manner. This could take place by means of trapped electrons as discussed in the second section.

The fact that partial space charge neutralization takes place even when no effort was made to accomplish it indicates that neutralization could be made very much more effective by introducing electrons by any of a number of special ways.

It must then be concluded that there is at present no indication that space charge is limiting the current that can be collected by this method.

Extension of Apparatus

In order to collect large amounts of material and extend the separation to large masses, the modifications necessary are clearly indicated. Extremely large ion sources can be built and the apparatus extended indefinitely in the axial direction repeating the unit shown in Fig. 11. For heavier masses the diameter of the apparatus must be enlarged several fold and higher voltages and fields must be used. On the basis of the work reported here, the radial separator appears to have unusual promise as a large scale large mass separator.