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A REVIEW OF SOURCE TECHNIQUES USED IN RADIOACTIVE ATOMIC-BEAM EXPERIMENTS

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ERRATA

To: All recipients of UCRL-9400  
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<u>Page number</u>	<u>Line</u>	<u>Change</u>
19	11	change " $\theta$ " to " $\phi$ ".
41	9	omit "H".
44	Sec. C-line 8	change "5/16" in. to "3/16" in.
44	last line	change "1/r" to "1/K".
48	line 12	disregard paragraph beginning "Marrus has ... "
60	line 1 (under <u>Bismuth</u> )	References are 57a, 57, 58.
62	line 21	change "band" to "hand".
67	line 10	insert ref. 68 after "Wexler".
68	line 1	insert ref. 69 after "Dodsworth".
69	line 15	change "90 $\mu$ A" to "120 $\mu$ A".
69	line 16	change "1000 $\gamma$ /hr" to "1000 r/hr".
69	line 21	insert ref. 72 after "Swift".
69	line 30	should read "water and 10 N sodium ..."
69	line 31	change (5) to (6).
70	line 2	should read "two drops of 10 N NaOH".
70	line 3	change "wave" to "wire".
71	4 under (Half Life)	change "34" to "39".
75	line 16	should read " $I^{124}$ (4 day)".
77	line 20	change " $O^{16}$ " to " $O^{15} O^{16}$ ".
77	last line	change "0.15" to " $O^{15}$ ".
82	line 2	change "though" to "through".

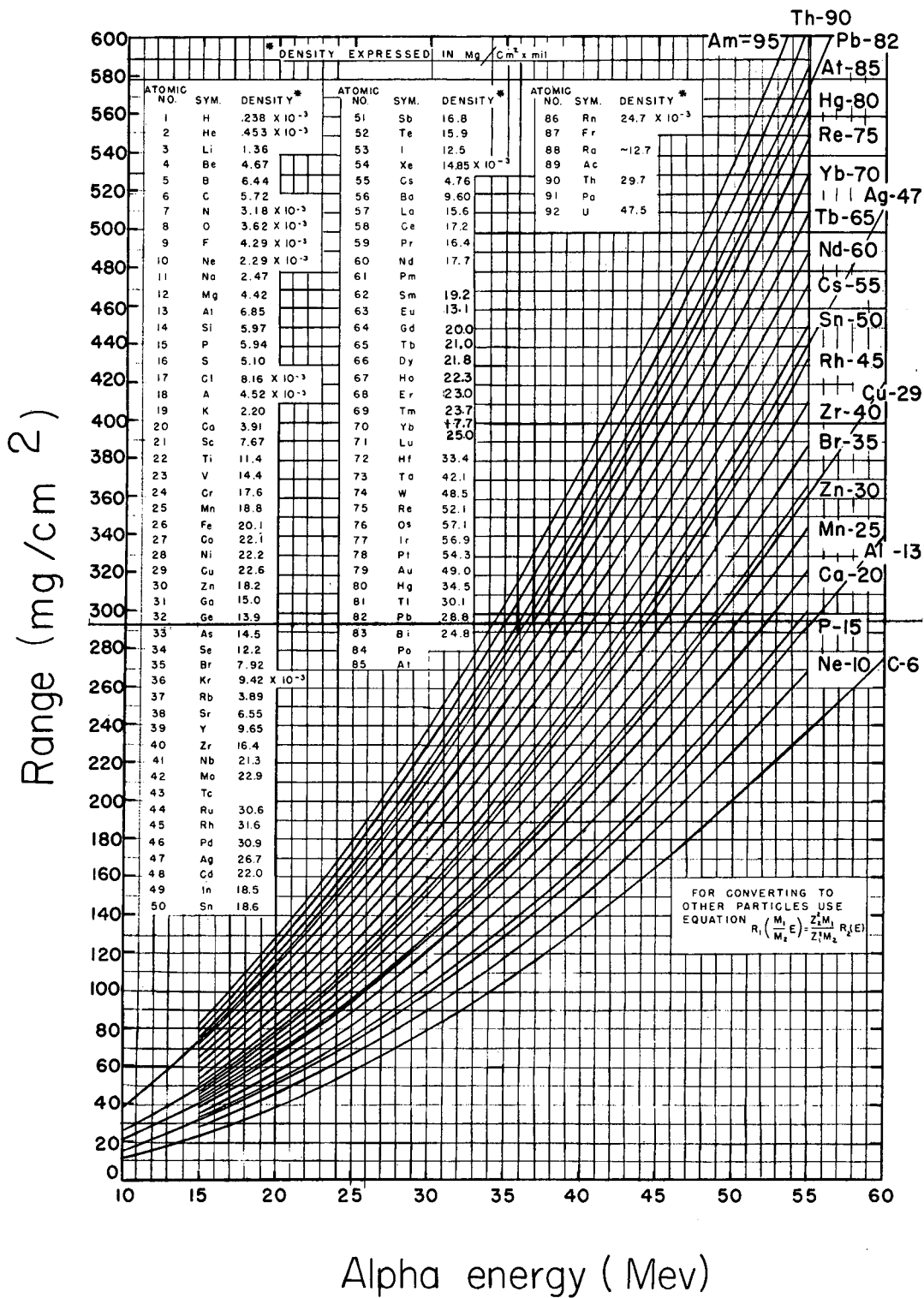
<u>Page number</u>	<u>Line</u>	<u>Change</u>
83	line 5	change "Ag <sup>105</sup> (8 day" to "Ag <sup>106</sup> (24 min)".
83	line 7	change "the same two isotopes" to "other silver isotopes".
88	line 19	should read "in a neutron flux of $5 \times 10^{10}$ neutrons/cms <sup>2</sup> /sec."
89	line 18	omit "of potassium and oxygen".
90	last paragraph	for "Te" read "Tl".
93	line 21	for "ground" read "around".
94	under Pa <sup>233</sup> (line 2)	read "decays to 27.4 day Pa <sup>233</sup> ".
96	line 3	change "thus" to "this".
97 through 103		remove Fig. 33 through 38. Insert attached Fig. 33, and new caption sheet (p.97).
105	Ref. 27.	correct spelling "Dostrovsky".
106	Ref. 44.	correct date "1960".
106	Ref. 45.	correct spelling "Nagle"
106	Ref. 48.	should read: "J. R. Zacharias and R. D. Haun, Massachusetts Institute of Technology Quarterly Report-34, October 1956."
106	Ref. 53.	should read: R. L. Christensen, Measurement of the Spin of As <sup>76</sup> by the Atomic-Beam Method (Thesis). Technical Report NYO-8016, Prince- ton, New Jersey."
106	Ref. 54.	should read: "Phys. Rev. Letters".
106	Ref. 56.	period after "Measurements". The LX-Rays of..."
106	Insert Ref. 57a.	Axensten, Johansson, and Lindgren, Arkiv Fysik 15, 463 (1959).
106	Ref. 59.	should read: Tl <sup>200</sup> , Tl <sup>202</sup> , In <sup>109</sup> , In <sup>110m</sup> , and In <sup>111</sup> .
107	Ref. 64.	change "Edwards" to "Edmonds".

<u>Page number</u>	<u>Line</u>	<u>Change</u>
107	Ref. 67.	should read: V. W. Cohen and D. A. Gilbert, Phys. Rev. <u>95</u> , 569 (1954).
107	Ref. 73.	correct spelling "Renzetti"
107	Ref. 76.	should read: Christensen, Hamilton, Lemonick, Pipkin, Reynolds, and Stroke, Phys. Rev. <u>101</u> , 1389 (1956).
107	Ref. 83.	should read: H. L. Garvin and E. Lipworth, Nuclear Phys. <u>19</u> , 140 (;960).
108	Ref. 84.	should read: Lipworth, Garvin, and Green, Phys. Rev. <u>119</u> , 2022 (1960).
108	Ref. 99.	change "668" to "686".
108	Ref. 103.	correct spelling "Transuranic". Omit "23" (date).
109	Ref. 110	Omit Ref. 110.

VII. APPENDIX

Fig. 33. Range Energy Curves for Alpha particles in various materials.

(Note conversion formula for other particles).



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Figure 33.



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A REVIEW OF SOURCE TECHNIQUES  
USED IN RADIOACTIVE ATOMIC-BEAM EXPERIMENTS

Edgar Lipworth

September 15, 1960

A REVIEW OF SOURCE TECHNIQUES  
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ABSTRACT

A description of specialized techniques used for the production of beams of radioactive atoms is presented. The production of isotopes by neutron, proton, deuteron, and alpha-particle bombardment is discussed. Special attention is given to practical details. Target holders, ovens, etc., and specialized equipment used for handling radioactive substances are described. Chemical techniques used in the preparation of individual isotopes are included.

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## I. INTRODUCTION

In recent years the atomic-beam method has been extensively used for studies of the electronic and nuclear properties of radioactive atoms. This article describes some of the specialized techniques that have been developed for the production of the beams themselves, i. e., the so-called source techniques. For those interested in a general description of the atomic-beam method and the results that can be obtained by its use, there now exist a number of excellent books and review articles to which reference may be made.<sup>1, 2, 3, 4</sup>

In atomic-beam experiments on stable isotopes, many and varied techniques have been employed to produce steady beams of atoms.<sup>5</sup> Beams have been produced in some cases by simply heating the element of interest in an oven, in others by allowing a compound of the element to react chemically with another element in an oven so that the first element is liberated in a controlled way. Atomic beams have been produced from molecular species by high-voltage direct-current or radio-frequency discharges (hydrogen, halogens).<sup>6, 7, 8</sup> Thermal dissociation has also been employed in the case of hydrogen,<sup>9, 10</sup> and sources have been developed for the production of beams of atoms and ions in excited (metastable) states.<sup>11-14</sup> Despite their diversity, however, these methods have a common factor. They have been used almost exclusively for producing beams under conditions where the atoms are all of the same chemical type, and present in more than trace quantities. (For beams produced by a chemical reaction within the source chamber, the same

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statement applies to each chemical constituent of the source.) The peculiar difficulties that are encountered when beams of radioactive materials are produced stem from the extreme smallness of the quantities of radioactive isotopes involved, ( $10^{10}$  to  $10^{14}$  atoms), and because very often the isotope is contained within a matrix of a large quantity of chemically dissimilar material, from which it must be extracted.

Radioactive isotopes can be prepared for atomic-beam experiments, by one or another of the following four methods:

1. By neutron bombardment of an isotope of an element either in natural or compounded form to produce another active isotope of the same element by the  $(n, \gamma)$  reaction
2. By bombardment of an element (combined or uncombined) with alpha particles in a cyclotron to produce an isotope of an element differing by two in nuclear charge by the  $(\alpha, kn)$  reaction ( $k$  integral)
3. By proton bombardment through the  $(p, kn)$  and  $(p, kp)$  reactions
4. By deuteron bombardment through the  $(d, kn)$  and  $(d, kp)$  reactions.

In the case of an isotope produced by the  $(n, \gamma)$  reaction in a stable chemically pure metal, no unusual difficulties arise. As the radioactive isotope is chemically identical to the stable isotope, the problem reduces to that of producing a beam of the stable isotope. If, for example, one desires to produce a beam of radioactive silver --  $\text{Ag}^{110m}$  (253 day) -- one would take a piece of silver foil or wire and place it in a pile until the desired specific activity was obtained and then place the irradiated material in the oven of the atomic-beam apparatus. In some cases, however, the element to be bombarded can be placed in the pile in only compound form. For example, it would be quite difficult to put liquid bromine inside of a pile and satisfy the safety regulations governing pile operation, but it is an easy matter to introduce  $\text{KBr}$  powder. Suppose one wants  $\text{Br}^{82}$ : In the pile some of the  $\text{KBr}^{81}$  is converted to  $\text{KBr}^{82}$  and the problem of producing a beam of  $\text{Br}^{82}$  is identical with that of producing a beam of  $\text{Br}^{81}$  from  $\text{KBr}^{81}$ . In practice, the bromine is liberated from the  $\text{KBr}$  by chemical methods, and the specific activity of the sample can be controlled by adding additional bromine; but the point remains that though a chemical operation was necessary to prepare the sample, the element whose beam is wanted is at all stages present in more than trace quantities.

The following factors govern the choice of the amount of parent isotope bombarded and the time of bombardment:

- (1) The neutron activation cross section.
- (2) The isotopic abundance and cost of the parent isotope.
- (3) The half-life of the daughter isotope.
- (4) The size of the oven slit.
- (5) The cost of bombardment.
- (6) The ease of handling.
- (7) The apparatus running time.

Although the extent of permissible variations in a given set of bombarding conditions is large, and a specific choice must depend upon an individual's judgment, some general remarks can be made. If the half-life of the daughter isotope is of the order of hours (say 1 to 12 hours) and the available quantity of activity is marginal, or if the parent isotope is expensive, then in order to make the best use of the active material, the sample should be effused from the oven in a time about equal to a half-life. Because there are definite limitations on the rate at which material can be effused from an oven with an exit aperture of given area, to produce an atomic beam (see Sec. III and VB) the amount of material irradiated will depend to a large extent upon the oven slit dimensions. If the parent isotope is cheap but the sample specific activity limited, this remark is still valid but no longer relevant or important if the specific activity is adequate for the experiment. If the half-life of an isotope is long (several days or weeks), it is usually economical to bombard several pieces of the parent material simultaneously and use them in successive runs.

The problems encountered in preparing sources of isotopes produced by the  $(n, \gamma)$  reaction can be solved in most cases by a simple approach; it is principally when isotopes are produced by one of the latter three methods mentioned above that difficulties arise. Suppose, for example, one wants to make a beam of  $I^{123}$  (13 hr). One way of producing  $I^{123}$  is to bombard antimony metal with alpha particles in a cyclotron. The reaction proceeds in two ways,  $Sb^{121}(\alpha, 2n)I^{123}$  and  $Sb^{123}(\alpha, 4n)I^{123}$ , the latter branch requiring higher energy than the first, and at the end of the bombardment, trace quantities of  $I^{123}$  ( $\sim 10^{12}$  atoms) are to be found trapped in about a gram of powdered antimony metal. If the antimony is placed in an oven and heated, the active material does not effuse in a steady manner but comes out in bursts as the temperature

is raised; also the activity is not effused in atomic form (it is not deflected by the inhomogeneous magnetic fields) but presumably as an antimony iodine complex.

Another example is the case of  $\text{Rb}^{81}$  (4.7hr) which has been produced by bombarding  $\text{KBr}$  powder with alpha particles; the reaction is  $\text{KBr}^{79}(\alpha, 2n)\text{Rb}^{81}$ . Again, the rubidium does not effuse in atomic form in a stable manner. There are some exceptions to this rule: For example, the silver and gold isotopes  $\text{Ag}^{103}$  and  $\text{Au}^{196}$ , produced by bombarding rhodium and iridium foils with alpha particles and protons respectively, effuse from the foils in atomic form and in a controlled way when the foils are heated in tantalum ovens.

Thus, although it is not universally true that atomic beams cannot be produced by simply heating the target when the parent and daughter isotope are chemically dissimilar, it is usually true, and resort has to be made to the techniques of radiochemistry in order to separate the active isotope from its parent. In principle, the method is simple. The target material is put into solution, stable carrier material chemically identical with the active isotope is added, and separated by chemical reaction from the main body of material. In practice, difficulties arise because of the intense radioactivity of the sample; many operations must be performed by remote control and the chemistries must be simple and rapid. Each isotope presents a different problem.

The sections that follow immediately are devoted to a discussion of the factors that govern isotope production by neutron, proton, alpha particle, and deuteron bombardment, and to a description of some of the experimental source and target arrangements that have been successfully used in atomic-beam experiments with radioactive atoms. The latter part of this article contains detailed descriptions of some of the chemical and physico-chemical techniques that have been developed to prepare the active material for the oven.



## II. THE PRODUCTION OF ISOTOPES BY PARTICLE BOMBARDMENT

### A. Neutron Bombardment

The activity of samples produced by neutron bombardment is controlled by adjusting the neutron flux and bombardment time. If the half-life of the daughter isotope is of some hours duration, the mass of target material is chosen so that it will effuse from the oven under some reasonable conditions for beam formation, in a time determined by the proposed length of the experiment. These "reasonable conditions" will, of course, be determined not only by the oven slit width and operating temperature but by the specific activity of the sample material.

The basic equation governing the production of isotopes by neutron bombardment is

$$N = \frac{\sigma F}{\lambda} N_0 (1 - e^{-\lambda t}). \quad (1)$$

Here  $N$  is the total number of product atoms formed by the bombardment of  $N_0$  parent atoms for a time  $t$  in a neutron flux  $F$ . The quantities  $\sigma$  and  $\lambda$  are the formation cross section and decay constant of the daughter isotope respectively; the decay constant  $\lambda$  is related to the half-life  $T$  by  $\lambda = \frac{0.693}{T}$ . A convenient form of this equation useful for rapidly estimating bombarding times is obtained by setting  $t$  equal to the half-life and expressing the resultant activity in millicuries. With a number of minor re-definitions the final activity expressed in millicuries is:

$$A = 8R\sigma x \left(\frac{m}{M}\right) 10^2 \text{ millicuries.} \quad (2)$$

Here  $R$  is the neutron flux in units of  $10^{13}$  neutrons/cm<sup>2</sup>/sec,  $m$  is the mass,  $M$ , the molecular weight of the parent isotope,  $\sigma$  the cross section expressed in barns and  $x$  the isotope abundance, expressed in percent, of the parent nucleus in the target. Neutron capture cross sections are tabulated conveniently in a number of places and are available these days for most elements of interest. 15, 16, 17

B. Proton, Deuteron, and Alpha-Particle Bombardment

In order to estimate bombardment times, particle energies, and beam currents required for the production of a certain quantity of radio-isotope, it is necessary to know the cross section for the reaction that produces the isotope, but as very few absolute measurements of total proton, deuteron, and alpha particle reaction cross sections occur in the literature one usually relies on rough theoretical estimates. In this part we discuss briefly the general features of nuclear reactions induced by protons, deuterons and alpha-particles in sufficient detail to indicate how estimates of the reaction cross sections are made.

At bombarding energies below 20 to 30 Mev per incident nucleon, nuclear reactions proceed by the amalgamation of the incident particle and target nucleus into a compound nucleus with all of the excess energy available as excitation energy. The excitation energy is dissipated commonly by the emission of one or more nucleons. The general features of reactions in this energy range are well understood.<sup>18, 19</sup> The reactions are characterized by a rapid rise of the cross section, for the emission of a particular group of particles, as the bombarding energy is raised above the threshold, followed by a rapid decrease as the energy is further increased and competition from other reactions at higher energies becomes appreciable.

The great majority of cyclotron-produced isotopes that have been studied in beam experiments to date are the neutron-deficient isotopes produced by the boiling off of one or more neutrons following the absorption of a proton, deuteron, or alpha particle. The cross section,  $\sigma(x, kn)$   $k = 1, 2, 3, 4$  for the capture of particle  $x = (p, d, \alpha)$  as a function of the center-of-mass energy,  $E$ , can be approximated by the product of two factors.

$$\sigma(x, kn) = \sigma_c [x(E)] \left\{ \begin{array}{cc} 0 & [1 - \gamma_{K-1}(E)] \\ [1 - \gamma_K(E)] & \gamma_{K+1}(E) \end{array} \right\} E_K \quad \begin{array}{l} E \leq E_K, \\ E_K \leq E \leq E_{K+1}, \\ E \geq E_{K+1}. \end{array} \quad (3)$$

where

$$\gamma_K = \left\{ \begin{array}{l} \left[ 1 + \frac{E - E_K}{\theta_K} \right] \exp - \left( \frac{E - E_K}{\theta_K} \right) \quad k \neq 0, 1 \\ 0 \quad k = 0, 1 \end{array} \right.$$

Here  $E_K$  is the threshold energy of the  $(x, kn)$  reaction in the center-of-mass system of coordinates and  $\theta_K$  is the nuclear temperature.

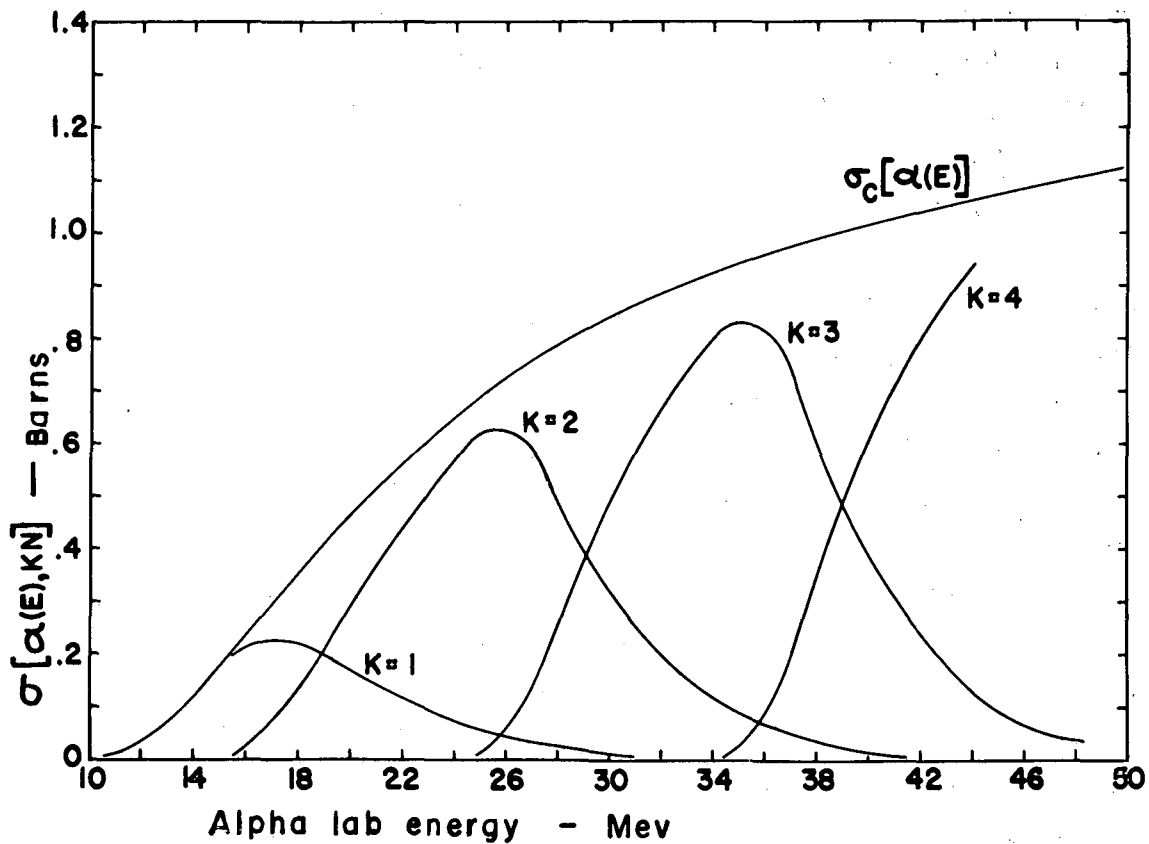
The first factor,  $\sigma_c [x(E)]$ , represents the cross section for the formation of the compound nucleus by bombardment with particle  $x$ ;  $\sigma_c$  is determined by the Coulomb barrier of the target nucleus. The second factor describes the decay of the compound nucleus as due to the evaporation of neutrons. The compound nucleus is considered to behave statistically as a highly degenerate fermi gas sufficiently excited to allow evaporation. The emission of  $k$ -neutrons by a nucleus with excitation energy  $E - E_1 > E_K$  is described as the successive evaporation of neutrons, each neutron leaving the nucleus excited with energy  $> E_1$  until  $k$ -neutrons are emitted. Here  $E_1$  is the energy required to boil off one neutron. For a particular excitation energy,  $E - E_1 > E_{k-1}$ , the compound nucleus depending upon the energy carried away by the first neutron may emit  $k, k-1, \dots, 1$  neutron(s) with the relative probabilities approximated by the second factor in Eq. (3).

The Coulomb factor  $\sigma_c$  has been calculated with continuum theory, by Blatt and Weiskopf and the results for protons and alpha particles are tabulated in Table 4.1 (Ref. 18, p. 352). Shapiro also has given tables of the Coulomb factor for protons, deuterons, and alpha particles and curves of the formation cross section versus energy.<sup>20</sup>

As an example of the curves that result from the above calculations, we reproduce in Fig. 1 the calculated excitation functions for  $\text{Br}(\alpha, kn)\text{Rb}$  reactions taken from the work of Doggett.<sup>21</sup> These curves have proved most useful in the calculation of yields in the intermediate mass range. The threshold energies  $E_K$  were obtained from mass spectrographic data and the known decay schemes. The nuclear temperature  $\theta_K$  was taken as 2.4 Mev, corresponding to an average excitation energy of 20 Mev.

The semi-empirical mass table of Cameron can be used to calculate reaction thresholds if the exact masses are not known.<sup>22</sup> At high  $Z$  values of the target nucleus, the Coulomb barrier is approximately 20 Mev for alpha particles so that production of  $k = 1$  isotopes is highly inhibited, the peak of the curve for single neutron emission occurring at 17 Mev.

Equation (3) gives the cross sections of reactions leading to excited states of the product nuclei. In regions of the periodic table where isomerism does not occur, these excited levels decay promptly into the ground state so



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Fig. 1. Calculated excitation functions for Br( $\alpha$ , kn) Rb reactions.

that the Eq. (3) represents the cross sections for the production of the product nuclei in their ground states. However, when isomerism exists, the compound nucleus can decay to an isomeric state of the product nucleus, and a knowledge of the decay of the isomeric level and its production relative to that of the ground state is necessary before the ground state cross section can be properly estimated.

### C. The (dp) Reaction

The (dp) reaction has been used on occasions to produce isotopes for beam experiments. Peaslee, using a semiclassical model, has calculated (dp) reaction cross sections; he assumes that a stripping process is responsible for the entire cross section.<sup>23</sup> In all cases the cross section is zero at a threshold ranging from 1 Mev in the case of Na<sup>24</sup> to 6.5 Mev for Th<sup>232</sup>, rising rapidly to a peak and then falling slowly because of competition from the (dn) reaction, which becomes important when the deuteron energy becomes comparable with the Coulomb barrier height.

In Table I we list a number of measured (dp) cross sections compiled from Peaslee's article. The cross sections and energies correspond to the maxima of the experimental curves, except in the cases of Au<sup>197</sup> and Th<sup>232</sup>, where measurements have not been made at energies high enough to develop the peak. In any particular case a rough idea of the peak (dp) cross section can be obtained by extrapolating between these few experimental points given in Table I.

Table I

Tabulation of peak cross sections for (dp) reactions on various elements. For Au<sup>197</sup> and Th<sup>232</sup> the cross sections are observed cross sections at the stated energies but the peak has not yet developed.

Target	Product	Energy (Mev)	Peak Cross Section $\sigma$ (barns)
	Na <sup>24</sup>	6	0.47
	Co <sup>59</sup>	8	0.27
	Cu <sup>63</sup>	8.2	0.22
	Br <sup>83</sup>	8.5	0.4
	Bi <sup>209</sup>	12	0.14
	Au <sup>197</sup>	9	0.2
	Th <sup>232</sup>	9	0.6

#### D. Other Reactions

A few radioisotopes have been produced in sufficient quantity for atomic-beam experiments by reactions other than those already discussed. For example, Davis<sup>24</sup> has made Na<sup>22</sup> by the (da) reaction on magnesium: Mg<sup>24</sup>(da)Na<sup>22</sup>; and Petersen et al.<sup>25</sup> have produced millicurie amounts of K<sup>43</sup> through the (ap) reaction on argon gas: A<sup>40</sup>(ap)K<sup>43</sup>. Also, recently Ewbank and Chan have made Ag<sup>113</sup> by the (ap) reaction on palladium: Pd<sup>110</sup>(ap)Ag<sup>113</sup>.<sup>26</sup> However, these reactions have not been extensively employed because the Coulomb barrier has to be penetrated twice and for medium and high Z nuclei the total reaction cross section is lower than when neutrons alone are emitted. In addition, there appears to be no simple theory described in the literature to enable one to make satisfactory estimates of the cross sections, although recently computer programs\* have been developed that make use of Monte Carlo calculational methods to find a particular reaction cross section and its energy dependence.<sup>27</sup>

#### E. Range Energy Curves

It is often advantageous to maximize the production of a particular isotope by adjusting either the target thickness (if this is a foil) or the energy of the incident particles. For example, referring to Fig. 1, if it were desirable to produce a target enriched in Rb<sup>82m</sup> (6.3 hr) and Rb<sup>84</sup> (33 day) produced by the (an) reaction on Br<sup>79</sup> and Br<sup>81</sup>, this could be accomplished by reducing the energy of the alpha particle beam (48 Mev at Berkeley) to about 18 Mev. This can most easily be done by introducing aluminum degrading foils in front of the target holder. For completeness, in the Appendix we include range-energy curves for protons, deuterons, and alpha particles in various materials, that should be useful.

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\* Apparently, the people at the Weizmann Institute, Israel, where these programs were developed, will perform calculations of reaction cross sections and their energy dependence for interested parties on request.

### III. SIGNAL STRENGTH

Here we discuss the factors that determine the number of curies of active material required for an atomic beam experiment. The following assumptions are made:

(1) The active material is effused from the oven in a manner such that the carrier material satisfies the conditions for molecular flow through the oven slit.

(2) The active material is ultimately mixed with the carrier and the beam intensity is uniform for the effusion time,  $T$ .

(3) The beam is deposited upon collector plates with collection efficiency  $\phi$ .

(4) The resonance peak counting rate is a fraction,  $r$ , of the full-beam counting rate. The full-beam rate is defined as the counting rate observed with all deflecting magnets turned off and the stop wire removed.

(5) The counter efficiency is  $\epsilon$  and the decay is simple, i. e., every disintegration of a nucleus on the collector plate is counted with efficiency  $\epsilon$ .

When atoms effuse from an aperture under conditions of molecular flow, it is easily shown that the ratio of the number of atoms effusing per second into a solid angle  $d\omega$  at angle  $\theta$  to the plane of the slit, to the total number effusing per second is equal to<sup>1</sup>

$$\frac{dn}{n} = \frac{d\omega}{\pi} \cos \theta.$$

So if we define  $dQ$  as the number of curies of activity striking the collector in time  $t$ , and  $Q$  as the total activity effused in time  $T$ , then

$$dQ = \frac{Qd\omega}{\pi} \left( \frac{t}{T} \right),$$

where  $d\omega$  is now the solid angle subtended by the collector at the oven.

The counting rate (per minute) of this collector is then

$$c = 2.2 \times 10^{12} Q \left( \frac{d\omega}{\pi} \right) \left( \frac{t}{T} \right) \phi \epsilon r,$$



where we have used the fact that 1 curie  $\equiv 3.7 \times 10^{10}$  disintegrations/sec.

As an example, consider the case of  $\text{Br}^{82}$  (35 hr) which has been studied at Berkeley.<sup>28</sup> The  $\text{Br}^{82}$  decays with the emission of a 0.46-Mev  $\beta^-$  particle followed by some  $\gamma$  rays. It is available in 100-mc lots from Oak Ridge National Laboratory as KBr powder. By the time the sample has arrived in the laboratory and has been converted to liquid bromine, the activity, because of decay and inefficiencies in the chemistry, has been reduced to perhaps 50 mc. During the experiment, the bromine vapor is dissociated in a discharge tube whose efficiency is 0.8, in an apparatus where  $d\omega = 0.5 \times 10^{-5}$  and  $r = 0.02$ . The bromine atoms are collected on freshly evaporated silver surfaces for which  $\theta = 1$ , and counted in low-background continuous-flow beta counters with an efficiency of 0.5 (half the decaying atoms send their  $\beta$  particles into the collector). In a typical run, the collection time  $t$  is 10 minutes and the effusion time  $T$  about 5 hours. The expected counting rate is therefore

$$c = 2.2 \times 10^{12} \times 0.05 \times \left( \frac{0.5 \times 10^{-5}}{\pi} \right) \times \left( \frac{10}{300} \right) \times 1 \times 0.5 \times 0.02 \times 0.8,$$

$$= 46 \text{ cpm.}$$

Resonances of about this magnitude are, in fact, observed above an apparatus background counting rate of about 10 cpm. This latter background arises from a slight unavoidable contamination of the collector buttons themselves (1 to 2 cpm), and from the diffusion of stray and scattered atoms and molecules within the atomic-beam apparatus. The  $\beta$ -counters in current use at Berkeley have background counting rates of about 3 cpm, and it is therefore quite easy to distinguish these resonance peaks above background with reasonable statistical uncertainty, in counting periods of 5 to 10 minutes. In any event, a reasonable rule of thumb to bear in mind is that in order to perform an experiment on an isotope decaying by  $\beta$  emission, about 10 to 100 mc of activity must be available. Whether more or less activity is required depends upon such factors as the fraction of the beam returned to the collector on resonance (this depends upon the mass and temperature of the beam atoms, and the complexity of the atomic system, particularly the number of fine and hyperfine structure states); the apparatus background, which varies from element to element; the local room background, and the general state of the apparatus.

For isotopes that decay by K capture, it is possible to build thin crystal scintillation counters to count the x ray that follows the K capture, which have a background of less than 1 cpm.<sup>29</sup> In this case, the requisite amount of activity can be somewhat reduced. Some of the heavy elements, e.g., plutonium, curium, americium, decay with the emission of alpha-particles for which counters are available with backgrounds lower than 1/10 cpm. Because the apparatus background with these heavy refractory elements is quite low, successful experiments have been performed on them with as little as 1 to 10 mc of activity in the oven.

The above remarks apply to atomic-beam machines that employ Rabi magnets; in machines with focusing magnets, the amount of activity required can be reduced perhaps by as much as a factor of 20, although the apparatus background with focusing machines appears to be relatively higher than in the more conventional type.

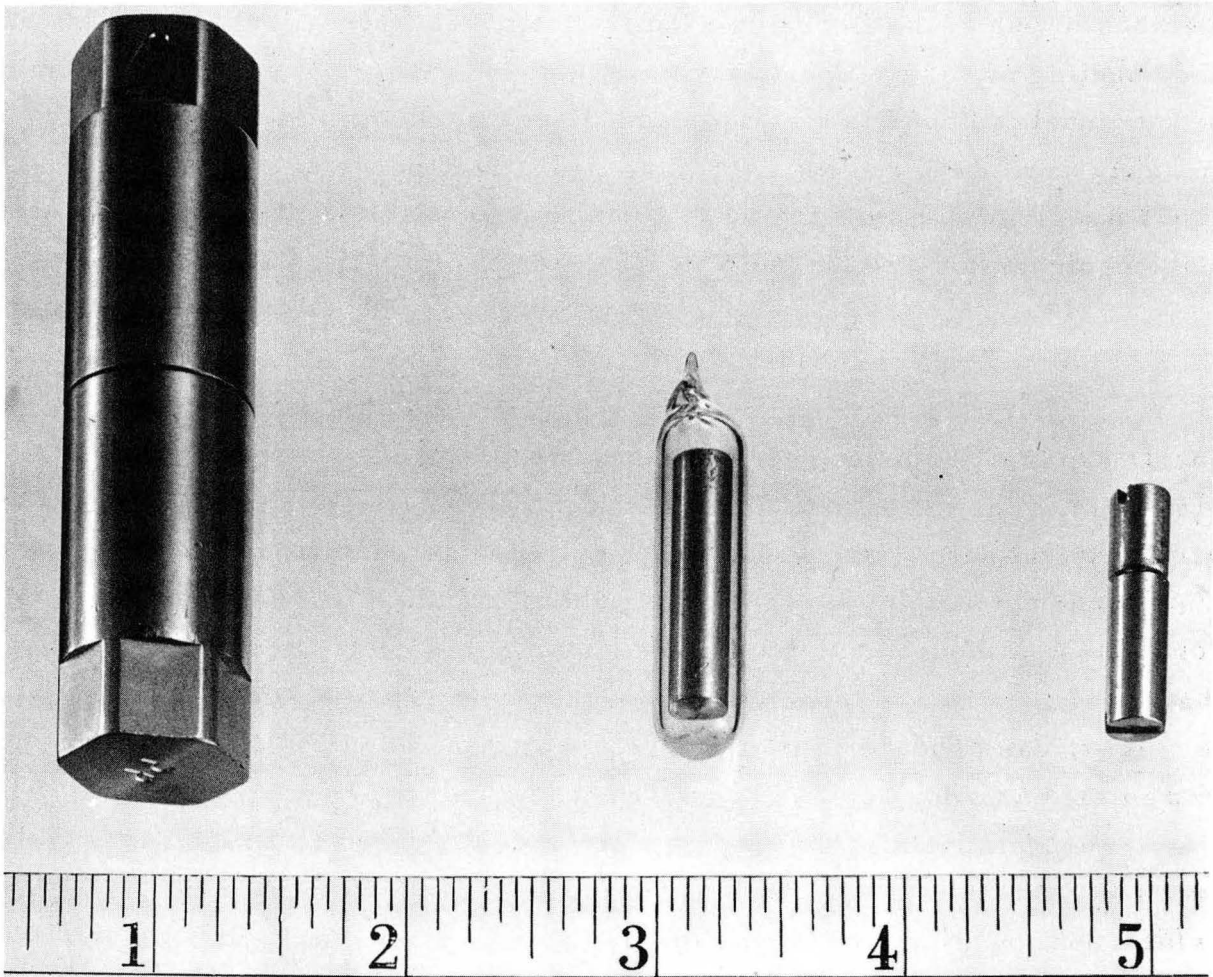
#### IV. ISOTOPE PRODUCTION-PRACTICAL DETAILS

The following sections give a detailed description of some of the methods in use at the Berkeley Atomic-Beam Laboratory for the production and handling of radioactive isotopes. The best source of general references to individual isotopes is the Table of Isotopes by Strominger et al. in which an exhaustive survey of the literature is neatly condensed.<sup>30</sup> There are two books now available that are quite good general introductions to the problems and techniques involved in handling radioactive isotopes.<sup>31, 32</sup>

##### A. Sample Holders for Neutron Bombardment

During a bombardment, the target material is sealed in a glass or quartz capsule protected by a container made of electrolytically refined aluminum. The latter material generates no long-lived activities under bombardment and is, consequently, much used as a containing material when high fluxes are employed, to minimize handling difficulties. Glass primary containers, although easier to seal than quartz, suffer from the disadvantage that the  $\text{Na}^{24}$  activity, generated from the neutron bombardment of  $\text{Na}^{23}$  in the glass, can produce radiation levels that make handling of the container extremely hazardous unless special precautions are taken.

Metallic targets become warm when exposed to a high neutron flux and it is advisable to blow the air out of the capsule with helium or argon before sealing it to prevent oxidation. This is particularly important with some of the more active rare-earth metals. After bombardment, the capsule can be crushed and the target material picked out with tongs, but a more sanitary procedure is to scratch the glass or quartz beforehand and break the tube in half in a special jig. Powdered targets, e. g., potassium bromide, should be rendered anhydrous before they are sealed into a capsule to prevent the possibility of a burst inside the pile. In Fig. 2 is shown a package used by Winocur for the bombardment of americium-241 where a dangerous level of alpha activity is encountered.<sup>33</sup> Approximately 5 mg. of americium oxide is contained in the smaller aluminum capsule which is sealed into the quartz tube as shown. The quartz tube is packed into the larger capsule with quartz wool plugs to prevent breakage.



ZN-2593

Fig. 2. Capsule used for bombardment of  $\text{Am}^{241}$  where a dangerous level of alpha activity is encountered.

### B. Sample Holders for Cyclotron Bombardments

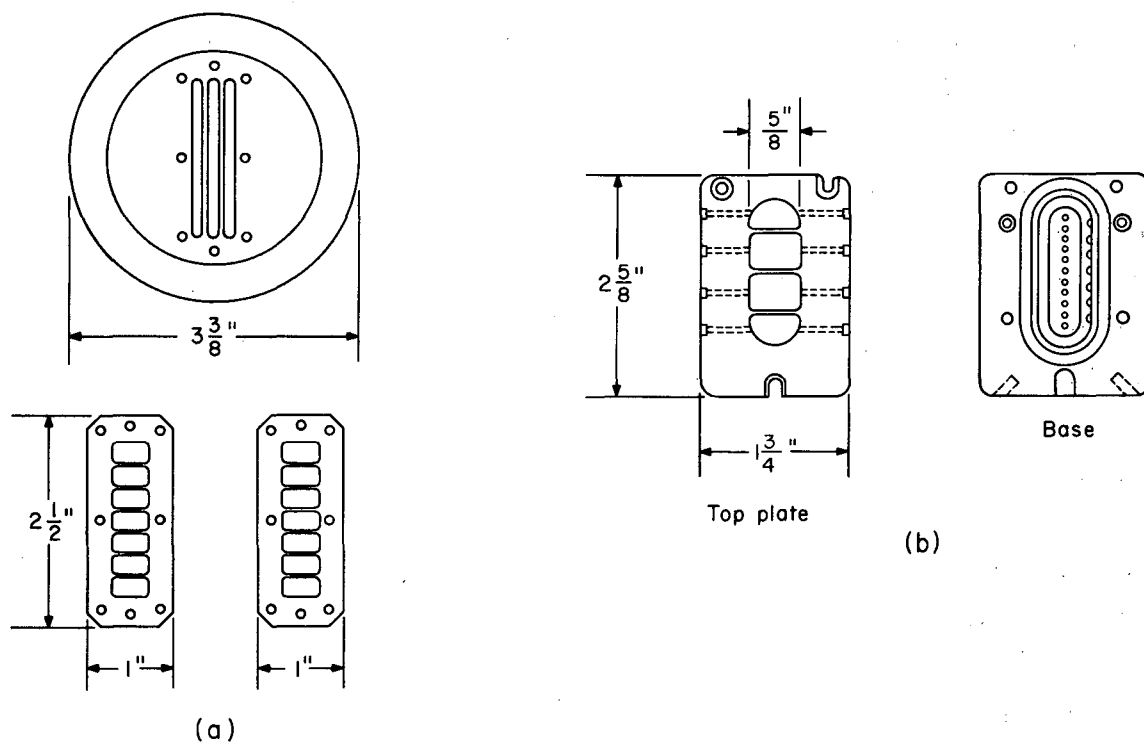
Figure 3 shows two sample holders that are in common use at Berkeley; the holders are again fabricated from electrolytically refined aluminum to minimize the radiation hazard. In type (a) the three grooves are each about 1/8 in. wide and 2 in. long and only a thin aluminum vane separates adjacent grooves. The groove depth varies from 0.010 to 0.050 in. depending on the desired target thickness. The target, usually in the form of anhydrous powder is packed into the grooves and covered with a 0.001 in. Al foil and held in place by one of the ribbed clamps. A second foil, held in place by the second clamp, minimizes the chance of scattering active material into the cyclotron should the first foil burst. During bombardment, the back of the holder is cooled with water. These holders have been extensively used for alpha particle bombardments at 40  $\mu$ A of beam current at an energy of 45 Mev; their chief disadvantages are that they take time to disassemble and it is a little awkward to remove the powder.

The holder shown in Fig. 3(b) has proved more convenient. A photograph of it is shown in Fig. 4. The target material is contained in a "boat" made by stamping a depression in a 0.010-in. thick Al foil the back of which seals against the O-ring. The foil is held against the O-ring by the ribbed clamp which can be removed in a matter of seconds simply by loosening the three screws the lower screw is hinged and flips back allowing the plate to be removed. The target material is usually covered by 1 or 2 sheets of 0.001-in. Al foil which are cooled by blowing helium across the front surface through holes drilled into the clamping plate. The back surface of the "boat" is cooled with a spray of water. These holders can quite safely be used with beam currents up to 25  $\mu$ A; at higher currents the foils tend to burn out more readily than in the holder described above, probably because of poorer cooling of the target material.

In Fig. 5 is shown the rather elaborate supporting block which is used to contain the holders during bombardment in the external beam of the cyclotron.\* The cyclotron beam enters from the right, passes through the

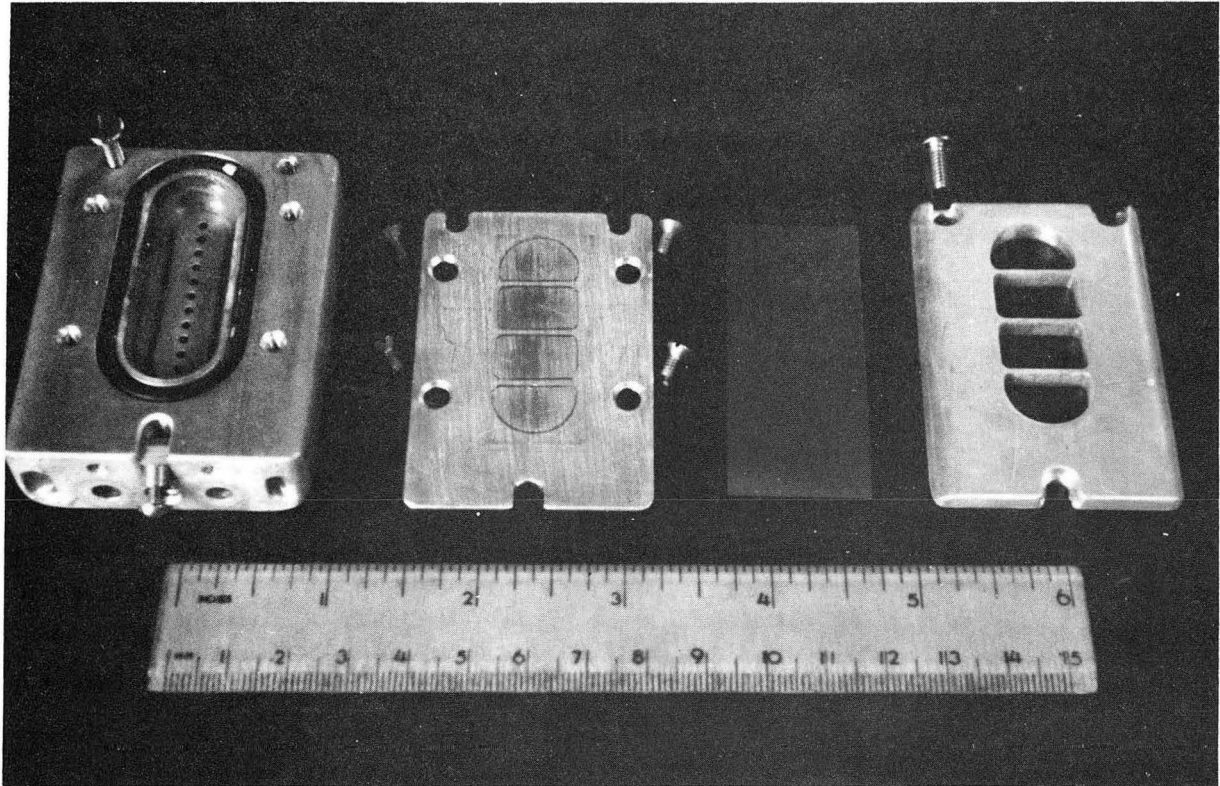
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\* I am indebted to Mr. R. Burton and Mr. B. Jones for this drawing of the cyclotron target-block holder used at the Crocker 60-inch cyclotron, Berkeley.



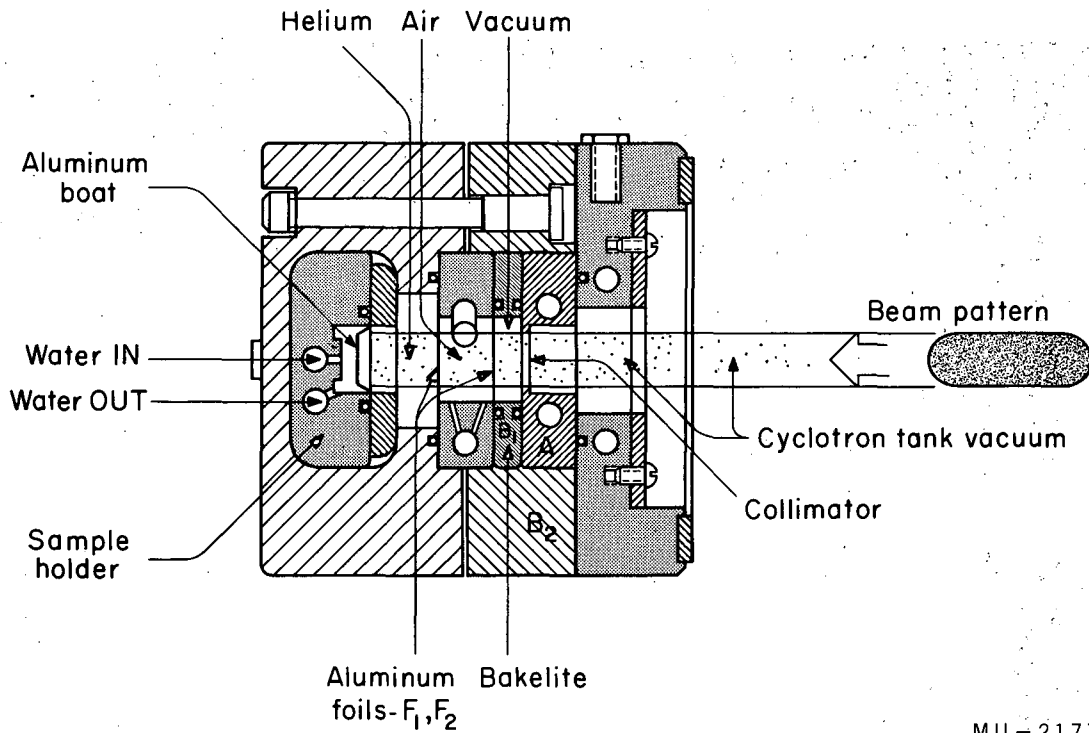
MU-21775

Fig. 3. Sample holders used for cyclotron bombardments of powders and foils.



ZN-2193

Fig. 4. Photograph of "Cats Eye" cyclotron bombardment sample holder used for powders and foils.



MU - 21774

Fig. 5. Supporting block for sample holder used during cyclotron bombardment

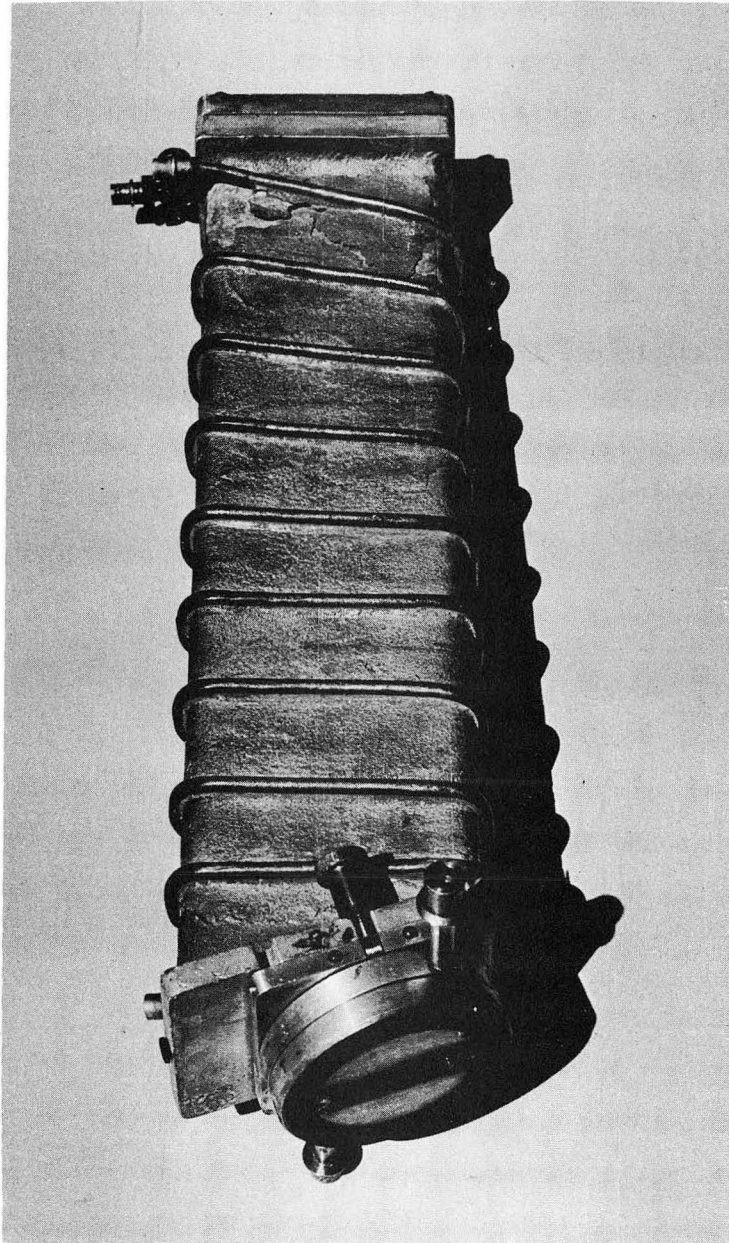


collimating slot and foils F1 F2, and falls upon the front of the target holder. The space between foils F1 F2 is filled with circulating air at a pressure of about 8 lbs/in.<sup>2</sup>; these foils isolate the cyclotron vacuum. The foil thickness determines the beam energy at the target and the circulating air keeps them cool. The space between the front of the target and foil F1 is filled with circulating helium at a pressure of 8 lbs/in.<sup>2</sup> whose function is to cool the front foil of the target holder. Should the foil break, the helium will not react with the hot target material. The pieces labeled B1 B2 are made of Bakelite and the rest of the block is made from electrolytically refined aluminum. The Bakelite pieces insulate the right-hand portion of the block containing the collimator from the left-hand part that holds the target; the ratio of the beam currents falling on the target and intercepted by the collimator can, in this way, be measured independently and used to monitor the bombardment.

The Berkeley 60-in. cyclotron is capable of producing accelerated particles with energies of 12 Mev per nucleon, at beam currents up to 40  $\mu$ A. The cross-sectional dimensions of the beam at the collimator are approximately 1/16-in.  $\times$  3/4-in.; such a beam would, in the normal course of events, rapidly burn a hole in a 1-mil aluminum foil, but luckily the beam wanders in a random fashion and no one part of the foil takes the full brunt for very long. The upper limit of bombarding current before foil burnout becomes too troublesome is about 40  $\mu$ A; an effective method of spreading the beam without loss of particles would probably enable the currents to be raised considerably. The use of rotating targets has not been exploited, but the advent of cyclotrons with beam currents considerably higher than 40  $\mu$ A should stimulate their development.

If the sample is in the form of a sturdy metal foil, considerably higher bombarding currents than 40  $\mu$ A can, of course, be employed. The internal circulating cyclotrons beam is considerably more intense than the external beam and holders been designed to enable foils to be rapidly transferred via vacuum locks in and out of the Dee tank.

Figure 6 shows a gas target holder that has been used by Shugart for the proton bombardment of noble gases to produce alkalies.<sup>34</sup> Its external dimensions are 19  $\times$  5  $\times$  1-1/2-in. The container is made of cast aluminum and cooled by water circulating through copper tubing welded to the walls. The window assembly contains two 0.001-in. Al foils to separate the



ZN-1730

Fig. 6. Sample holder used to bombard gaseous targets in a cyclotron.

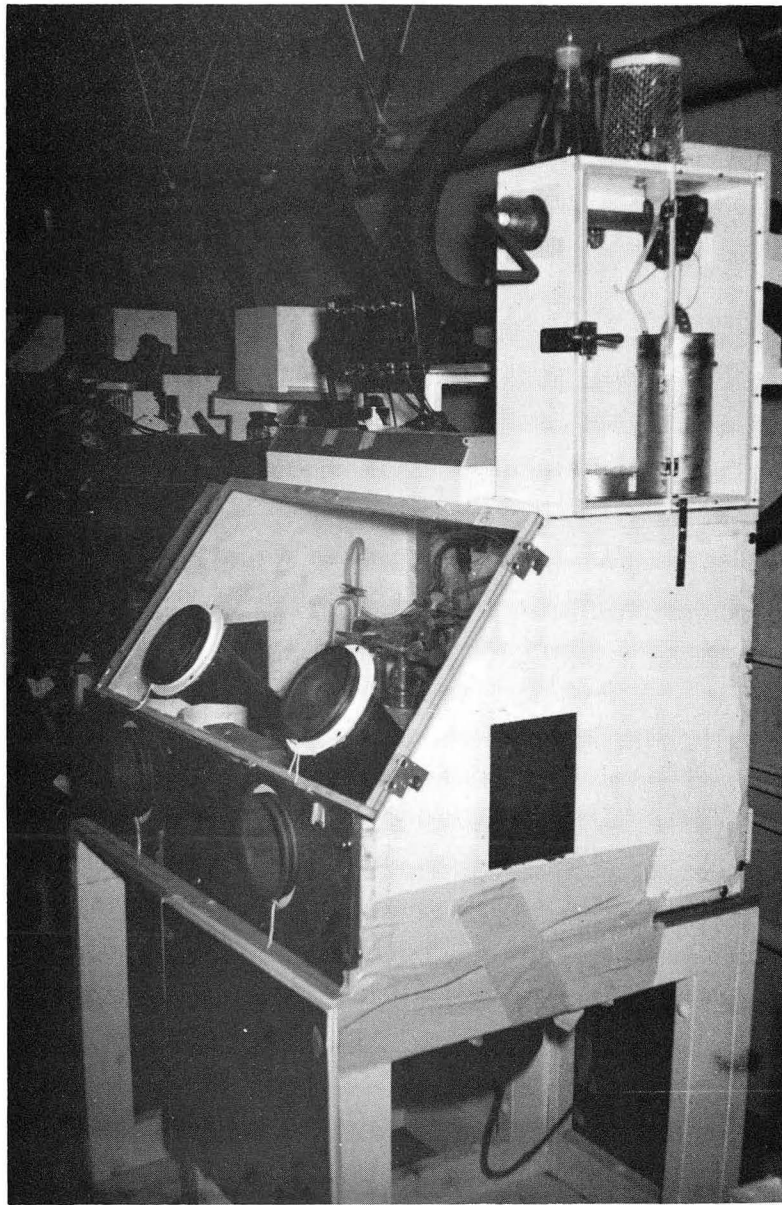
noble gas from the cyclotron vacuum. The space between the foils is cooled by a flow of air at 1/2 atmosphere pressure. For some details of its use see the later section on cesium.

### C. Boxes and Caves for Sample Chemistry

All chemical operations are best performed either inside a gloved box or "cave". The box (Fig. 7) is made of wood, lined with polyethylene sheet, and is held under a slight negative air pressure to prevent escape of spilled or volatile radioactive substances. All manipulations inside the box are performed through rubber gloves attached to the four ports, and materials and apparatus are passed into the box via sliding doors at the side. The boxes usually contain a centrifuge and electrical outlets, and compressed air, gas, and vacuum lines can be brought into the box at convenient points via filters. Inside the box, individual pieces of apparatus can be shielded by judiciously stacking lead bricks, and the front transparent surface can be covered with a lead glass shield to minimize radiation. Boxes are cheap enough to construct so that they can be thrown away once they have been contaminated; and although they are, in general, very safe to use, they have one weak spot in the rubber gloves, which tend to crack if not replaced periodically. It is wise to practice a particular chemical operation inside of a box several times with nonradioactive substances before attempting to process an active sample. This precaution can sensibly diminish the radiation exposure of the operator.

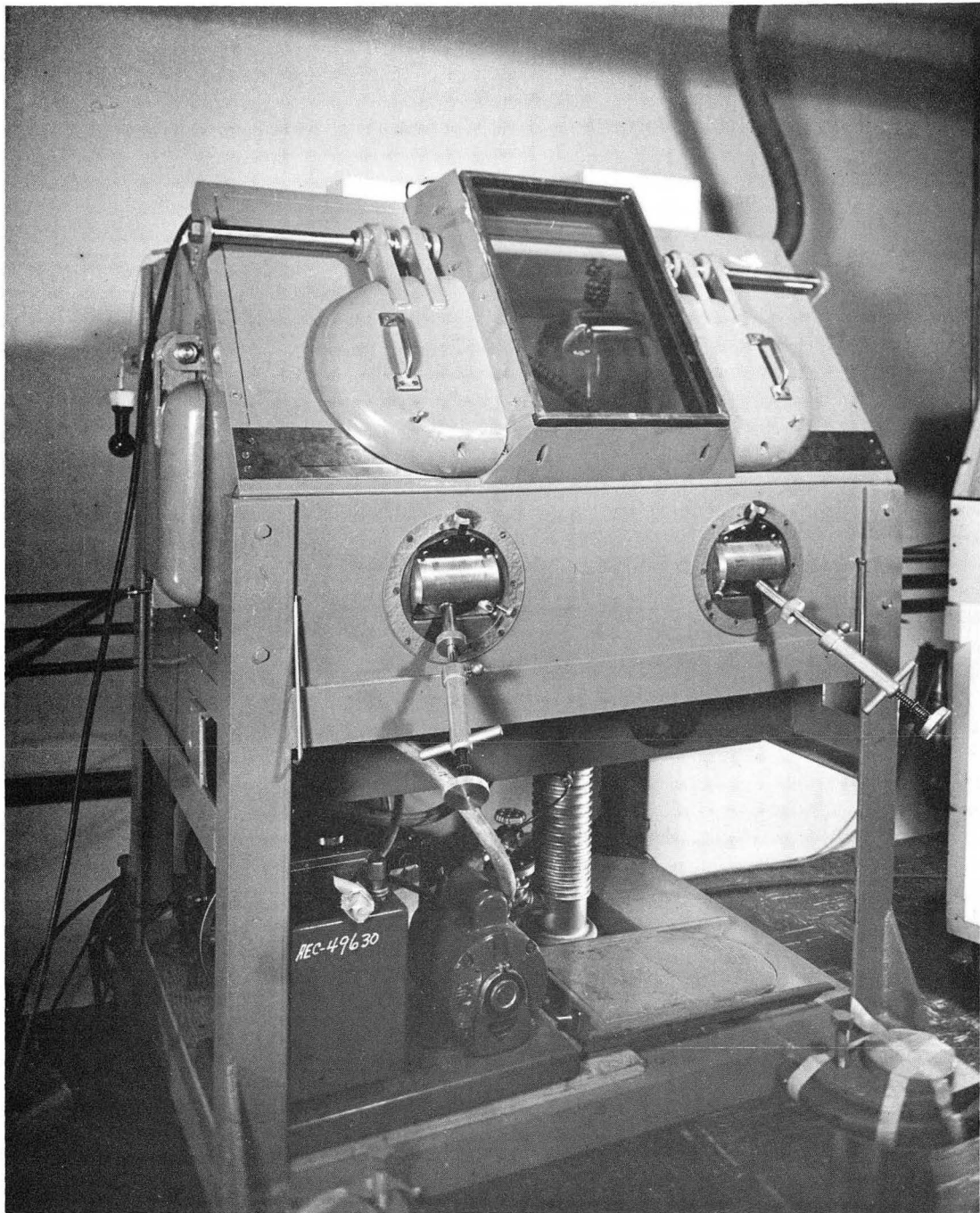
The cave (Fig. 8) is a lead structure inside of which a "box" is fitted. The cave walls are made of 2-in. thick lead plates and the ports can be covered by sliding lead doors. The cave is usually employed for handling only intensely radioactive sources or those requiring a long and difficult chemical processing. Apparatus in a cave can be handled by manipulators controlled from the outside, but in general, the use of manipulators wastes time. An operation that can be performed in ten seconds by hand may take several minutes with manipulators. It is obviously not good sense to use a cave if the operator's integrated exposure to radiation is greater with it than without it.

While it is true that working inside of a box or cave is rather inconvenient, it is definitely not recommended that chemical operations on volatile radioactive substances be performed in a conventional chemical hood.



ZN-2211

Fig. 7. Gloved box employed to handle radioactive materials.



ZN-2592

Fig. 8. Lead "cave" for handling highly active radioisotopes.

Recent experience indicates that draughts, even with a well-lowered hood are paradoxical in their behavior and can easily release active material into the laboratory.

## V. BEAM PRODUCTION

### A. Ovens and Discharge Tubes

The general principles that govern the production of an atomic beam are well understood and excellent discussions of this topic can be found in the books of Ramsey and Smith.<sup>35, 36</sup> For present purposes it is sufficient to note that in the case of molecular flow through an aperture (i. e. when the mean free path,  $\lambda$  of the effusing molecules or atoms is much larger than the linear dimensions of the aperture) the beam intensity at a distance,  $r$ , from a source of area  $a$  is:

$$I = 1.118 \times 10^{22} \left( \frac{ap}{r^2 \sqrt{MT}} \right) \text{ atoms/cm}^2/\text{sec},$$

where  $p$  is the source pressure in mm of Hg,  $M$  is the molecular weight of the emerging molecules and  $T$  the temperature (in  $^{\circ}\text{K}$ ) of the source. In most beam experiments the source is a slit in the wall of a heated oven or discharge tube, and the important condition that the above equation be valid is in practice that  $\lambda \gg w$ , the slit width. If  $p$  is increased until  $\lambda$  is comparable to or less than  $w$ , a cloud of molecules is formed in front of the slit which tends to inhibit further increase of intensity. In work with radioactive beams, the active isotope (for reasons discussed previously) is usually mixed with a stable carrier material which usually is, but may not be, chemically similar to the active isotope. In order that the optimum conditions for beam formation be met, the vapor pressure of the carrier isotope under operating conditions must be such that the condition  $\lambda \gg w$  is not violated. Obviously, a wide choice of operating conditions exist due to the control one has over the specific activity of the oven load.

The fraction of material emerging from an oven that falls on a detector at distance  $r$  can be increased by replacing the slit by a channel of width  $w$  and length  $l$ ;  $l$  is usually greater than  $w$ . If  $\lambda > l$ , the flow is molecular and the total amount of material effusing in a given time is reduced as compared to the case of a plane slit but the angular distribution of the emergent material is changed so that a greater fraction proceeds in the forward direction. A detailed discussion of channeled ovens is given in Sec. V-B but it can be remarked here that the use of channeled slits to conserve activity has

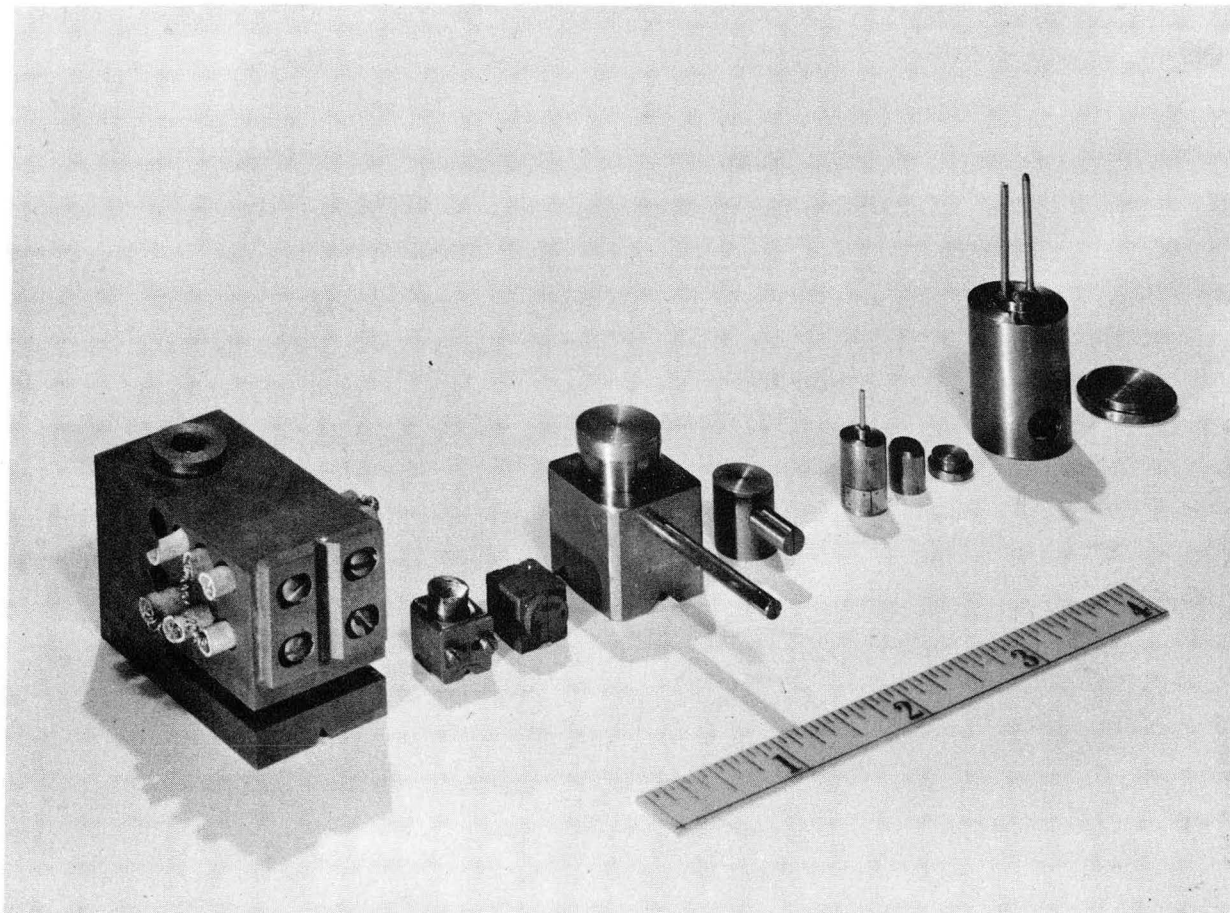
not been exploited as much as it might be in radioactive beam experiments.

Figure 9 shows typical ovens that have been used to produce beams. The ovens are heated either by coiled tantalum heater wires slipped into ceramic tubes that pass through holes drilled into the oven block, or by electron bombardment. Tantalum heaters have proved more serviceable than the more commonly used tungsten heaters, for tantalum does not become brittle and crack. Tantalum has been much used as an oven material at Berkeley, where much of the work has been concerned with beams produced at very high temperatures; but for many elements, ovens of stainless steel or iron can be used. Ovens have been successfully made from carbon with carbon slits.

Figure 10 shows an oven designed by Worcester to produce beams of gallium that can be very easily and cheaply produced. The carbon plug that contains the slit is held by a force fit into the cylindrical oven block.<sup>37</sup> For antimony and bismuth, which effuse in molecular form, a stainless steel block, fitted with a tantalum snout heated by electron bombardment has proved very successful (Fig. 9d). The snout is heated sufficiently to dissociate the antimony or bismuth molecules and the oven temperature is maintained by conduction of heat to the block down the snout; the snout length is determined empirically. The cylindrical ovens are supported in specially designed mounts (see Sec. V-D) on a thin tantalum support rod pressed into the base. This method of support minimizes the power required to obtain a given temperature by reducing conduction losses.

The ovens that have been employed to form beams of the transuranium elements are of interest. The need for high temperatures puts severe restrictions on the choice of materials from which to construct the oven, for it must not interact appreciably with the material under investigation at the temperature required to form a beam. Up to the present, the material which has been found most satisfactory is tantalum (melting point,  $3000^{\circ}$  C). Ovens made from tantalum have been used successfully in work on americium, curium, and neptunium. A tungsten oven with tantalum slits containing an inner liner of tungsten to control creep was found to be an excellent container for plutonium;<sup>38</sup> this oven is illustrated in Fig. 11. The hole is covered with tantalum slits made from 3-mil tantalum foil that is spot welded onto the front face of the oven. A sharp lip is ground into the inner liner to prevent creep of the plutonium. The outer shell is capped by a well fitting lid. The use of an open inner



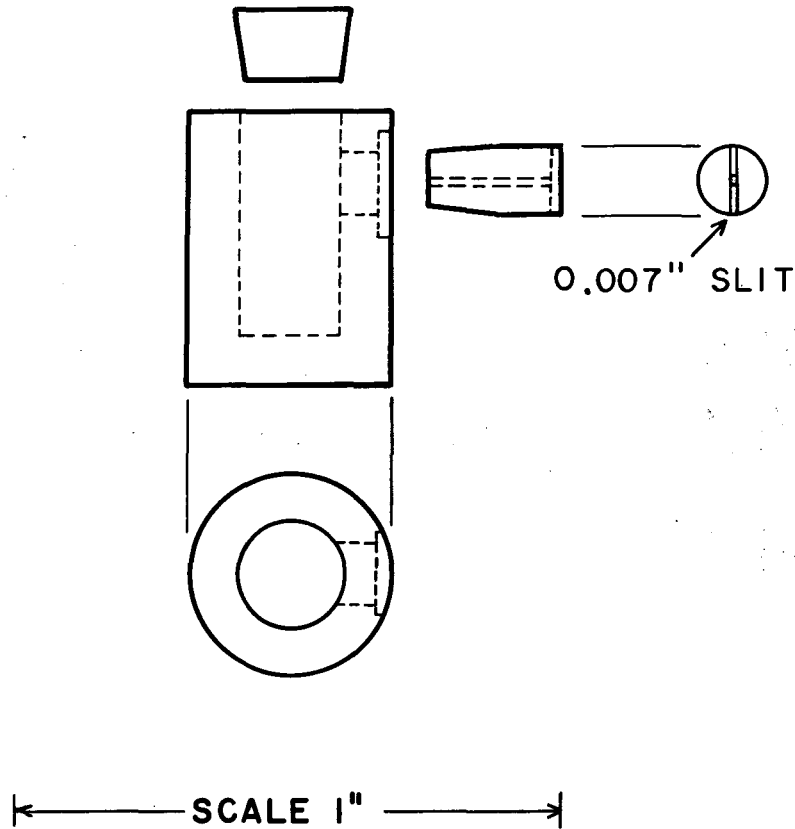


ZN-2594

Fig. 9. Various atomic-beam ovens. They are, left to right:

- a. Resistance heated stainless steel oven .
- b. Tantalum oven.
- c. Carbon oven.
- d. Dissociation oven.
- e. Channeled oven.
- f. Cylindrical tantalum oven with inner crucible.
- g. Large tantalum oven to produce tall beams (see text).

With the exception of oven (a), all ovens are heated by electron bombardment.



MU-13684

Fig. 10. Design for a simple carbon oven (see text).

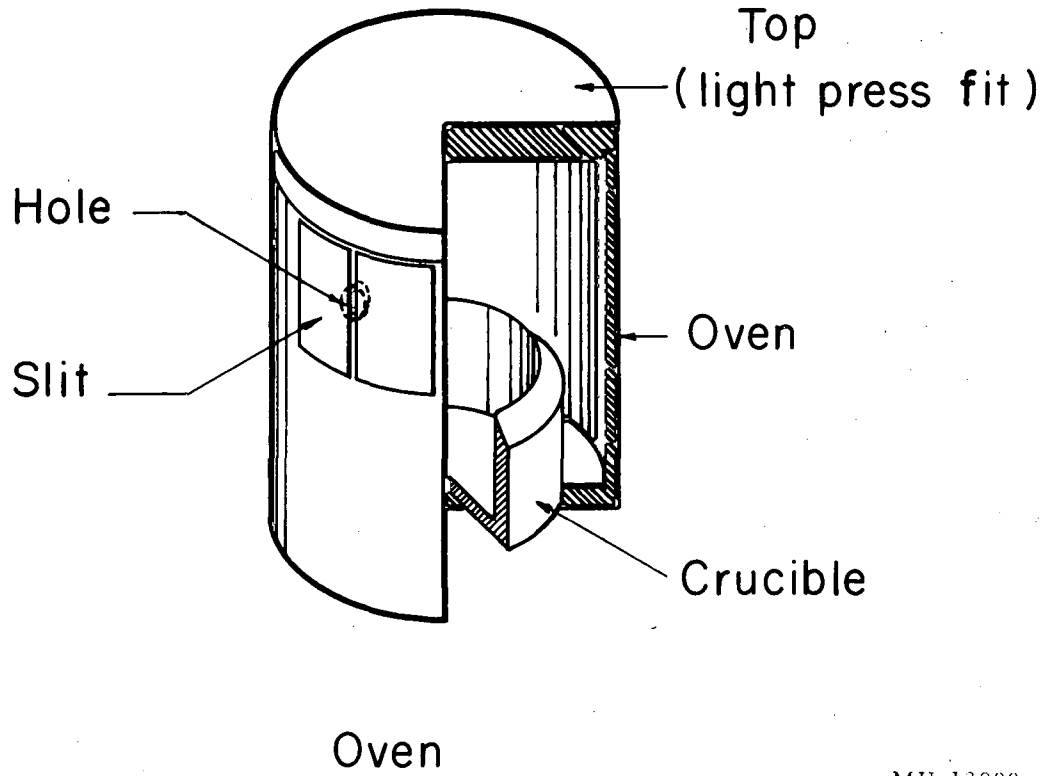


Fig. 11. Tungsten oven with inner liner used to produce beams of plutonium metal.

MU-13888

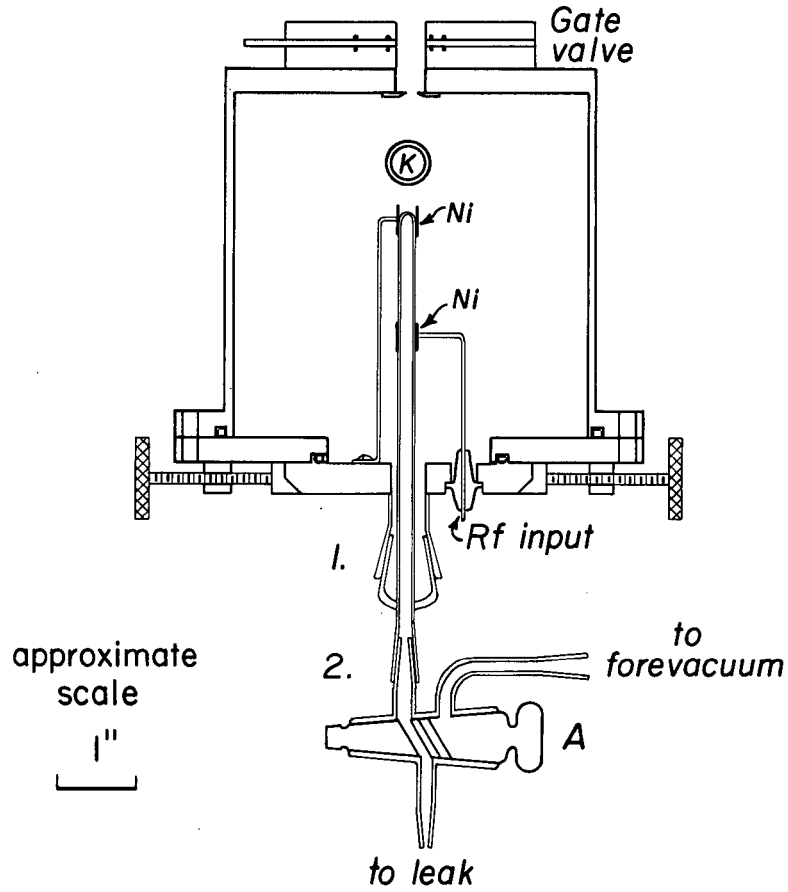
liner, with a sharp lip as a means of preventing creep, has had widespread application. Cabezas<sup>39</sup> has produced beams of most of the rare earth metals with no difficulty, using tantalum ovens with sharp-lipped tantalum liners, and it is now almost routine procedure to incorporate a liner in every oven.

Recently, Alpert has produced a beam of platinum by heating the metal in a closed carbon liner, itself contained within a closed tantalum cylinder.<sup>40</sup> The beam emerges through two slits, one in the carbon liner, the other in the container.

The oven shown on the extreme right in Fig. 9 is for use in an atomic-beam apparatus designed to pass broad tall beams. The oven slit is 1 in. high and 0.1 in. wide. It is hoped that by increasing the slit area in this way it will be possible to obtain increased signal intensities of highly refractory materials at lower temperatures. The oven is massive and is supported from two tantalum rods spot welded into the base.

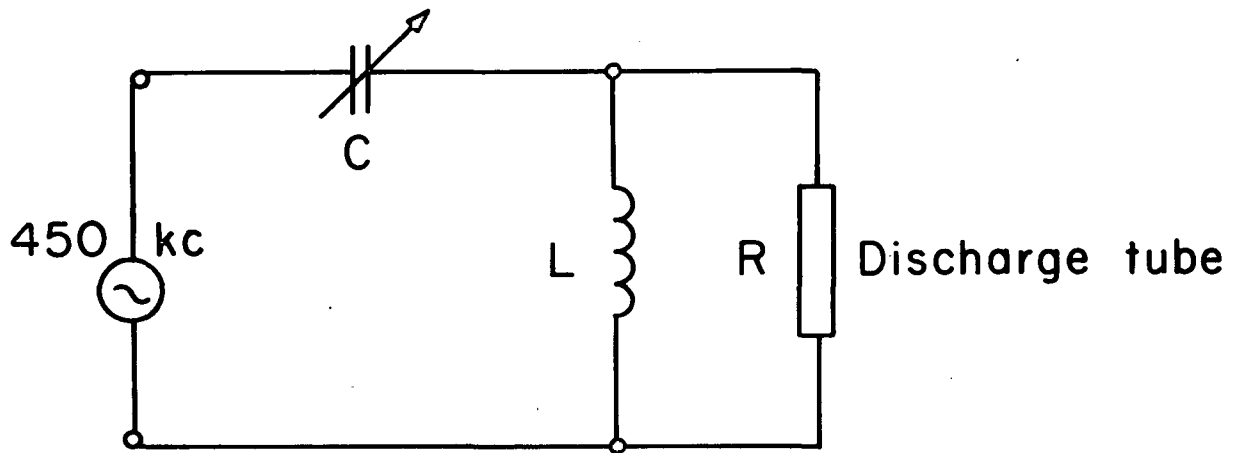
The halogens, bromine iodine and astatine, require special sources. The halogens exist as diatomic molecules and must be dissociated into atoms before they can be studied in an atomic beam experiment. Bromine and iodine have been successfully dissociated in the discharge tube shown in Fig. 12.<sup>41</sup> The tube is made of quartz, and the slit is cut into the front end, which has been blown thin, with a fine saw. The electrodes are nickel foils spot welded around the tube, and electrical contact is made by the nickel spring fingers. For efficient operation, it has been found essential to coat the tube under the electrodes with aquadag which presumably allows good capacitive coupling to the discharge. The tube is operated from a rf oscillator at 450 Kc. and a dissociation efficiency of 80 to 90% is easily obtained. The circuit shown in Fig. 13 is used to stabilize the current through the discharge tube.<sup>42</sup> It relies upon the little known property of a series resonant circuit that at resonance the current through  $R$  is independent of  $R$ . The discharge tube is connected across the inductance  $L$ , and when the circuit is operated slightly off resonance an extremely stable discharge results, one that has been run for hours without attention.

An even simpler source of iodine atoms is the thermal dissociator shown later in Fig. 23. Here the iodine vapor passes up a platinum tube approx 1/8 in. in diam., the end of which is heated to 750° C by electron bombardment. Dissociation efficiencies of 70% are easily obtained by this method. Bromine molecules cannot be dissociated readily by thermal means because of their high dissociation energy.



MU-17252

Fig. 12. Radio-frequency discharge tube used to produce beams of atomic iodine and bromine.



MU - 21836

Fig. 13. Radio-frequency discharge tube stabilizing circuit.

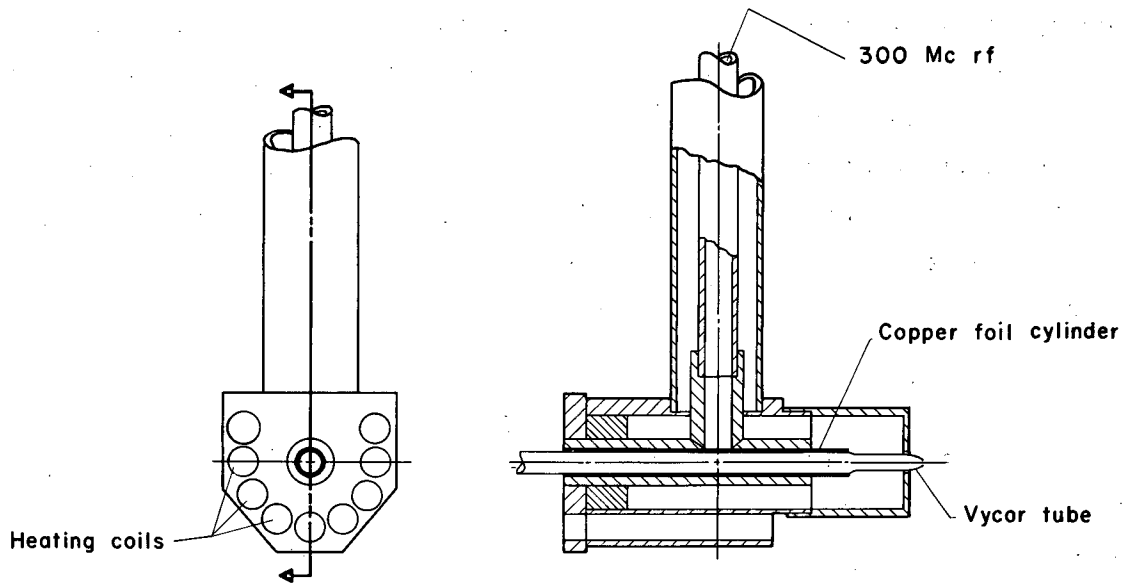
Astatine has presented some special problems and the source shown in Fig. 23 was built to solve them. The source resembles the iodine dissociator except that provision is made to heat the attached glassware to  $100^{\circ}$  C by an external heater and an iodine-astatine mixture was fed into the dissociation tube through a fritted glass leak. (See Sec. VI)

Christensen has used a heated discharge tube to produce an atomic beam of arsenic. The tube is shown in Fig. 14; the T section at the end of the coaxial line forms a resonant cavity which can be heated by heaters placed in the holes H running parallel to the length of the cavity. The discharge tube itself is simply a 3-in. long piece of 6-mm Vycor or quartz tubing with a snout at one end. Some results obtained with this tube are discussed later in the section on arsenic but it might be mentioned now that the tube operates at 300 to  $350^{\circ}$  C and produces a 25% dissociated beam of arsenic.

#### B. Channeled Ovens - Theory

It was noted in Sec. V-A that there is a way of making a more effective use of the oven-load by allowing the beam to effuse, not through a plane slit, but through a long, narrow channel. The effect of the channel is to modify the angular distribution of the emergent beam in such a way that its width, for the same forward intensity, is reduced below that obtained with the plane slit. Ramsey has collected together a number of formulae governing effusion from channels of different shapes and sizes which are reproduced here for reference.<sup>43</sup> The formulae are applicable to effusion from channels of length  $l$ , height  $h$ , width  $w$ , and radius  $r$ , when the pressure is sufficiently low for collisions inside the channel to be neglected, i. e., when the mean free path  $\lambda > l$ . If  $Q$  is the total number of atoms/sec effusing from the channel, and  $Q_0$  the number that would effuse from a plane source of the same cross sectional area as the channel, then for the same forward intensity we have

$$Q = (1/K) Q_0,$$



MU-21837

Fig. 14. High temperature radio-frequency discharge used to produce a beam of arsenic atoms.



where for

- (a) Any shape aperture of very short length or with  $l = 0$ ,

$$1/K = 1$$

- (b) A long cylindrical tube with  $l \gg r$ ,

$$1/K = 8r/3l$$

- (c) A long rectangular slit with  $h \gg l$ ,  $l \gg w$ ,

$$1/K = \frac{w}{l} \ln \left( \frac{l}{w} \right)$$

- (d) A long rectangular slit with  $l \gg h$ ,  $l \gg w$ ,

$$1/K = \frac{1}{lwh} \left\{ w^2 h \ln \left[ \frac{h}{w} + \sqrt{1 + \left( \frac{h}{w} \right)^2} \right] + wh^2 \ln \left[ \frac{w}{h} + \sqrt{1 + \left( \frac{w}{h} \right)^2} \right] - \frac{(l^2 + w^2)^{3/2}}{3} + \frac{l^3 + w^3}{3} \right\}.$$

- (e) A long rectangular slit with  $l \gg h$ ,  $l \gg w$ ,  $h \gg w$  (a special limiting case of (d) above),

$$\frac{1}{K} = \frac{w}{2l} \left\{ 1 + 2 \ln \frac{2h}{l} \right\}.$$

Recently, Giordmaine and Wang have derived expressions for the forward intensity  $I(0)$  and the half-width  $\theta_{1/2}$  of a beam effusing from a long tube under some quite general conditions, (when collisions within the tube are not neglected) that should prove useful to designers of channeled ovens.<sup>44</sup> They find

$$I(0) = \frac{3^{1/2} c^{-1/2} a^{1/2} (N)^{1/2}}{8 \cdot 2^{1/4} \pi^{1/2} \sigma}$$

$$\theta_{1/2} = \frac{2^{7/4} 3^{1/2} \sigma (N)^{1/2}}{1.78 a^{1/2} c^{-1/2}}$$

where  $\sigma$  = molecular diameter for collisions (cms),  
 $a$  = radius of tube (cms),  
 $N$  = rate of flow through tube, molecules/sec<sup>-1</sup>,  
 $\bar{c}$  = average molecular velocity, cms/sec<sup>-1</sup>,  
 $I(0)$  = beam intensity in forward direction, molecules (steradian)<sup>-1</sup>  
 sec<sup>-1</sup>,  
 $O_{1/2}$  =  $\frac{\text{beam half width}}{2}$ .

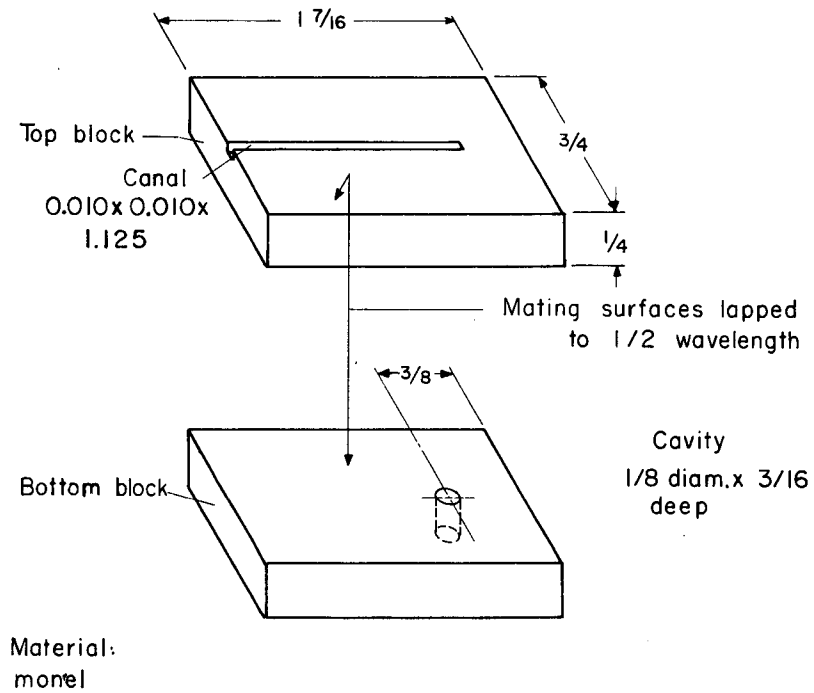
For a given forward intensity, the source length can be raised up to a critical length  $\Lambda$  beyond which the collimation does not increase. This critical length  $\Lambda$  is given by the expression:

$$\Lambda = \frac{2.5 \cdot 2^{1/4} \bar{c} a^2}{8 \pi^{1/2} \sigma^2 \cdot I(0)}.$$

### C. Channeled Ovens - Experimental

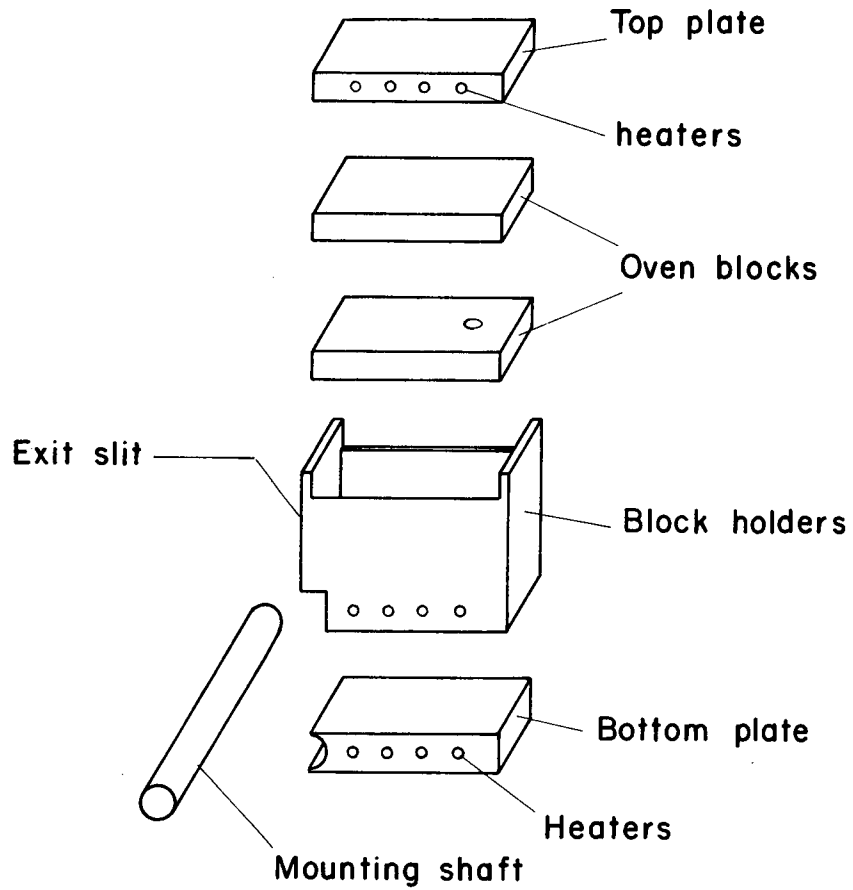
Only two types of channeled ovens have been successfully employed to date in experiments with radioactive atoms and these only with the radioactive alkali metals. The first, employed by Davis in work on Na<sup>22</sup> is shown in Figs 15 and in Fig. 16 is shown an exploded view of the oven blocks and their mount.<sup>45, 46</sup> The oven is fabricated from monel metal which possesses the smallest power of absorption of alkali metals. The oven channel is a slot 0.010 × 0.01 × 1.125-in. milled into one of the oven blocks. The oven well, 1/8 in. in diameter and 5/16 in. deep, is drilled into the other block in a position to overlap the interior end of the slot when the two pieces are mated together, and the two mating surfaces of the blocks are lapped flat to within .00001 in. to prevent leakage. The oven mount heaters are of molybdenum wire coated with aluminum oxide for insulation and the whole assembly is mounted on a shaft that can be rotated to obtain the rather critical alignment required with this type of oven.

The second type of channeled oven, employed by Stroke et al. in their work with radioactive cesium isotopes, is shown in Fig. 17.<sup>47</sup> The slits consist of nine 26-gauge hypodermic needles cut to 1/2 in. in length and silver soldered in a holder. The needles are flattened on two sides before mounting and 56% of the total slit area is open to the beam; the ratio 1/r



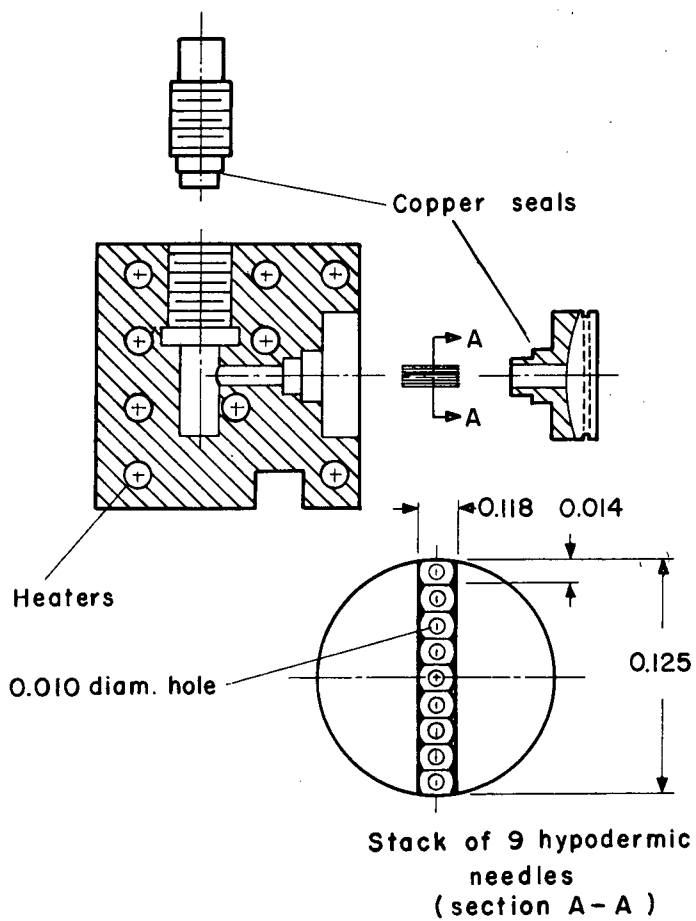
MU-21838

Fig. 15. Oven blocks. For channeled oven. All dimensions are in inches.



MU - 21839

Fig. 16. Holder for channeled blocks shown in Fig. 15.



MU - 21840

Fig. 17. Channeled oven employing stacked hypodermic needles as the channeled slits.

is approximately 100. The oven, block, plug, and slit holder are made of monel metal and the oven is heated by molybdenum wire coils insulated by thin quartz tubing. The sample is contained in monel metal cup and a set of baffles (not shown in Fig. 17) is screwed into the bottom of the plug to prevent "Spritz". The bottom of the plug, and needle holder, where they contact the oven block, is copper plated to obtain a soft metal seal against leakage. An iron-constantan thermocouple can be screwed into the oven block to measure its temperature.

Zacharias and Haun describe a source used to produce a well-collimated beam of cesium.<sup>48</sup> It is made by alternating layers of plane and corrugated nickel foils 0.001-in. thick, on top of one another.

Marrus has designed a channeled oven for use with the cyclotron-produced rare earths.<sup>49</sup> It is shown in Fig. 17. The channel consists of a 0.040-in. diameter hole, 0.375-in. long, drilled into a tantalum rod which is forced into the side of the oven block. The hole terminates before the end of the rod is reached and is continued by a slit. At the time of this writing no details of its performance are available.

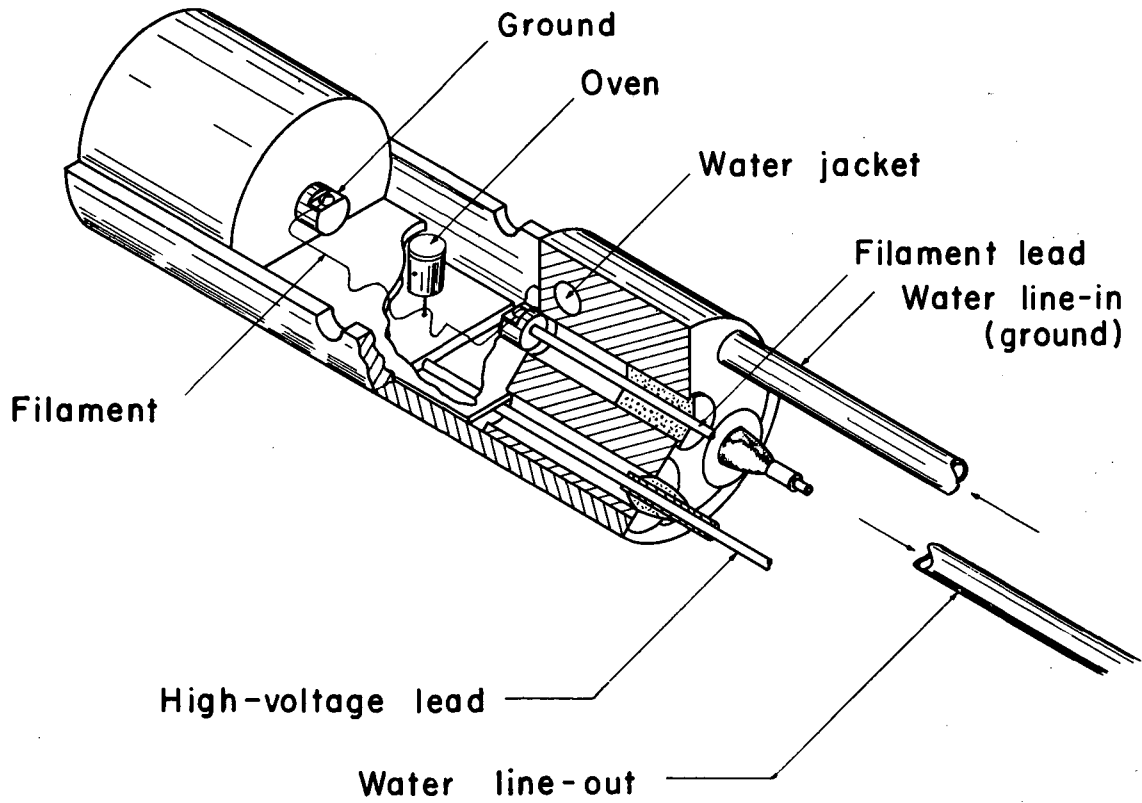
#### D. Oven Mounts

The oven mounts that have been used in radioactive beam experiments are quite elaborate and expensive devices requiring many hours of machine-shop time for their manufacture. Because the mounts rapidly become badly contaminated in operation and can only be handled inside of a gloved box, it is worthwhile to give considerable thought to their design in order that all adjustments and repairs can be easily made and that their life is as long as possible. A few general rules can be stated. All electrical feed-through insulation should be of the high-temperature ceramic type and should be hard soldered in place; the Kovar glass insulators tend to crack easily and cannot be replaced inside the box. The filament supports should be easily accessible and attention paid to the fact that filaments must be changed by hand and as rapidly as possible because of radiation hazard. All exposed insulator surfaces, particularly those carrying high voltage leads in electron bombardment oven mounts should be protected by removable shields to allow the insulators to be cleaned. The whole mount if possible, should be shielded to minimize the amount of material left in the apparatus oven can. An apparatus can be

dangerously contaminated if this precaution is not taken, particularly when long-lived isotopes emitting high-energy gamma rays, are being worked.

A type of oven mount that has been very successfully used for the past few years in the Berkeley Atomic Beam Laboratory is shown in Fig. 18.<sup>50</sup> The holder is designed so that it can be inserted and withdrawn from the apparatus vacuum system without bringing the whole apparatus up to atmospheric pressure, an important consideration in lengthening the life of the calibrating beam alkali metal, which is contained usually within the main oven chamber. The holder body is made of copper and is supported on stainless steel tubes which provide water cooling. The oven is mounted on a tantalum stem which fits into a hole drilled into the high voltage lead passing along the bottom of the loader. A removable plate effectively screens the high-voltage lead support insulators from being dirtied by oven material during operation, and a semicylindrical cap closes the whole loader to reduce contamination of the can. The beam escapes through a small hole in the side of the loader; a similar hole at the rear is to allow the oven temperature to be monitored. Figure 19 indicates how the loader is inserted into and withdrawn from the vacuum system. The upper view shows the loader ready to enter the machine. The plunger on the right is inserted so that the machine vacuum is sealed from the oven region, and in this position the marked pump-out is evacuated to mechanical pump pressure. After roughing down, the oven part is allowed to move further into the can until the cylindrical parts of oven and plunger unite. As the plunger is pulled back the oven part, under the action of the atmosphere, moves to its final position shown in the lower view; the plunger is finally pulled back to expose the oven as shown. This procedure is reversed in order to remove the oven from the system. The plunger is released and allowed to come into contact with the oven part; as the plunger moves in further, it pushes the oven part out, sealing the vacuum in the apparatus. Air is admitted through the pump-out and the oven part withdrawn completely. This loader operates horizontally however models that came through a top plate have been constructed and perhaps have some advantages.

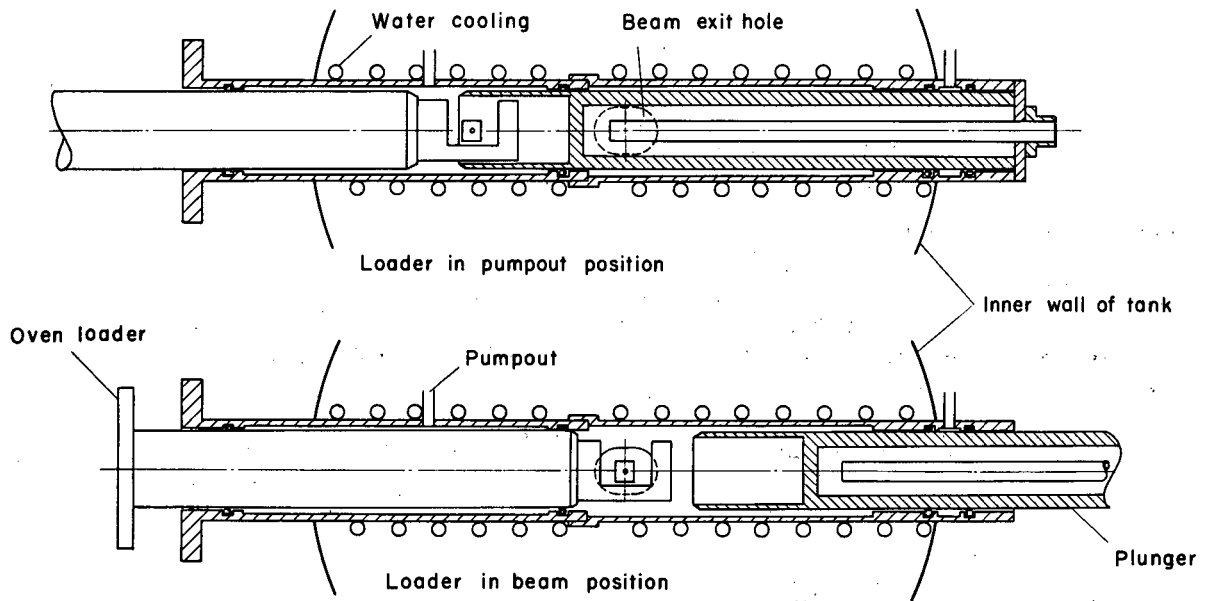
An end mounting vacuum loader used by Axensten et al.<sup>51</sup> is shown in Fig. 20. The oven is mounted on the end of a steel tube which is pushed into a water-cooled brass tube through a gate valve. Heating equipment,



MU-21841

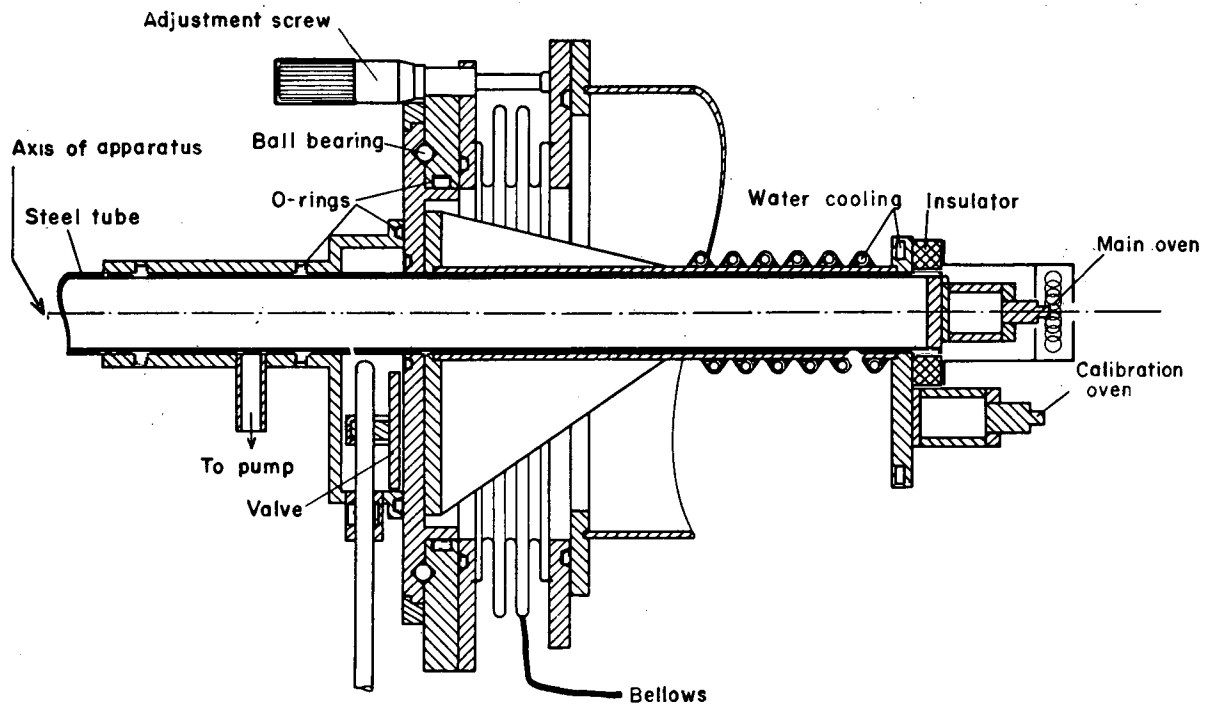
Fig. 18. Oven holder for rapid transfer of a loaded oven into an atomic beam apparatus. This holder operates horizontally and perpendicular to the beam direction.





MU-21842

Fig. 19. Schematic diagram showing operation of oven holder of Fig. 18.

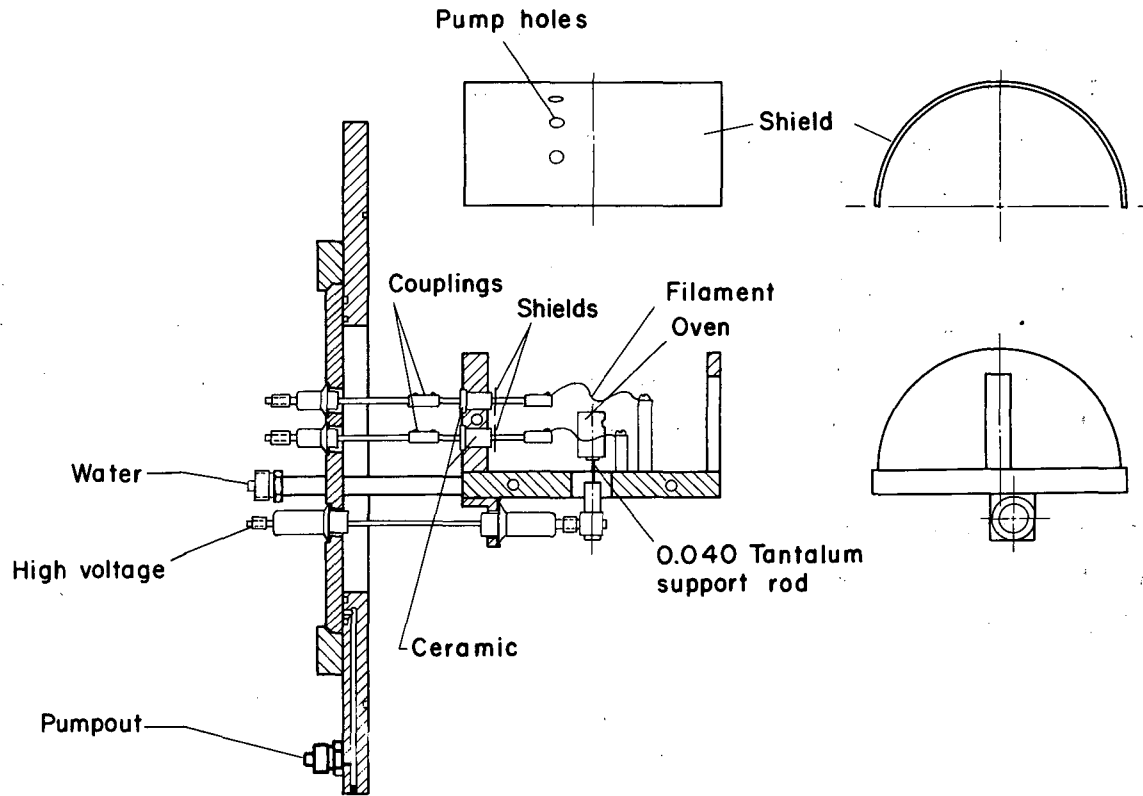


MU - 21156

Fig. 20. End mounting oven holder for rapid transfer of an oven into the apparatus vacuum.

radiation shields, and the calibration oven are mounted on a copper plate at the end of the brass tube. The whole assembly can be rotated on the ball bearings about an axis of rotation that permits either of the two ovens to be placed at will on the beam center line.

End mounting loaders of a type which do not pass a complicated vacuum lock have also been extensively used at Berkeley. In Fig. 21 a picture of such a loader, which is simple and sturdy, is shown. The large oven is supported directly from the high-voltage lead on an adjustable clamp. The interior insulating supports are removable, and are easily replaced if they become dirty. With this type of loader the oven can must be brought up to atmospheric pressure before the loader can be removed. As the calibration oven containing an alkali metal is also usually situated in the oven can and the alkali must be protected from attack by air, the can vacuum is released by bleeding in dry nitrogen and the machine repumped immediately. An alkali load can be preserved for several weeks if this precaution is taken; in newer machines the calibrating oven is contained within a separately pumped chamber which can be isolated by valves when the oven can is brought up to air.



MU-21843

Fig. 21. End mounting oven holder.

## VI. CHEMICAL TECHNIQUES EMPLOYED FOR THE PREPARATION OF INDIVIDUAL ISOTOPES

The following pages describe some of the specialized chemical and physico-chemical techniques that have been employed to prepare radioisotopes for the oven. The isotopes are catalogued alphabetically with the exception of the transuranium elements which are treated as a group under the heading "Transuranium Elements". A small section is given over to the rare earths; all of the rare earth isotopes studied up to the present have been produced by neutron bombardment, and beams of each produced in an identical fashion.

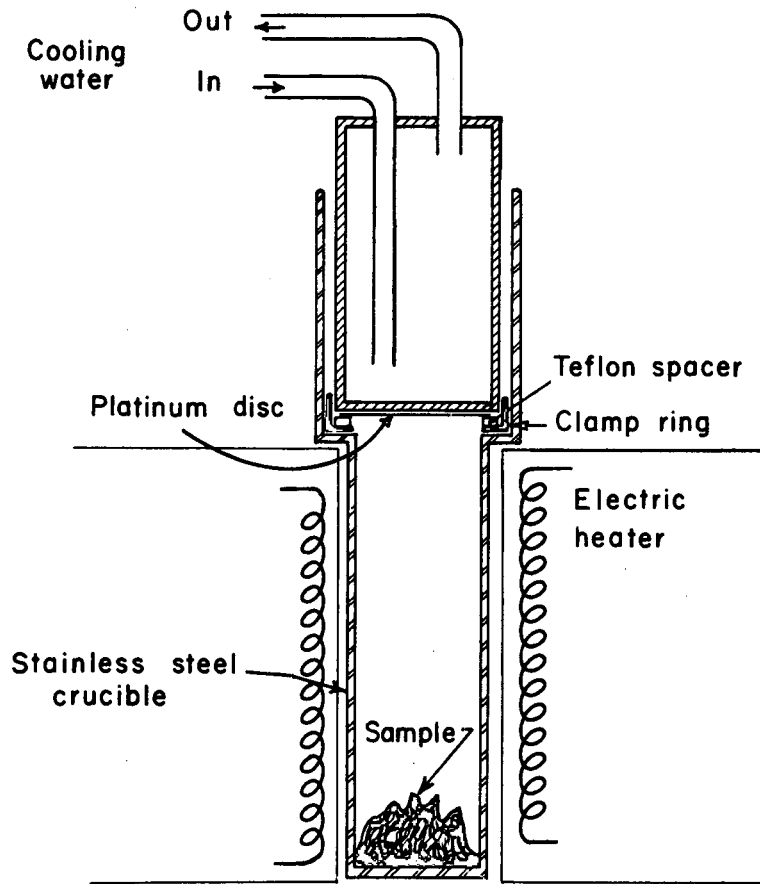
## 1. Arsenic

An atomic beam of  $\text{As}^{76}$  (26.5hr) has been made by Christensen. Arsenic tends to vaporize as polyatomic molecules which must be dissociated before an atomic beam experiment is possible and Fig. 14 shows the discharge tube assembly used to dissociate the arsenic; it has already been described in Sec. (V~~A~~). One end of the quartz tube is formed into a snout; the arsenic crystals are put into the tube through its back end which is then sealed. The tube is supported inside of a copper sleeve which fits snugly inside the coaxial T section and which provides a 1/4 in. gap over which the discharge voltage is developed. About 50 watts of heater power are needed to raise the tube to operating temperature (300-350°C), when the vapor pressure of the arsenic is about 0.1 to 0.2 mm of Hg. The discharge is maintained by driving the cavity with a magnetron oscillator and started by sparking the arsenic vapor with a Tesla coil. The fractional dissociation of arsenic atoms produced by the discharge is estimated to be about 25% but great difficulty was apparently found in maintaining a steady discharge and great variations in signal intensity required that a complicated normalization procedure be employed to make meaningful sense out of the results. The tube was filled with about 200 milligrams of stable arsenic, sufficient to maintain a discharge for 6 to 12 hours. The arsenic beam was detected by allowing it to fall on a copper disc cooled to liquid-nitrogen temperature. After a disc was exposed, it was removed from the apparatus and sealed in a matrix of moisture-absorbing paper and Scotch tape to prevent loss of condensate.

## 2. Astatine

In 1940 Corson, MacKenzie, and Segre isolated a radioactive element whose chemical, physical, and nuclear properties established it to be element 85, the last of the halogen group. This element, which does not possess a stable isotope, they named "astatine". The measurement of the nuclear spin of 7.2-hour astatine-211 by an atomic-beam experiment constitutes the first direct spin determination of an isotope having no stable counterpart, whose half-life measured in hours.

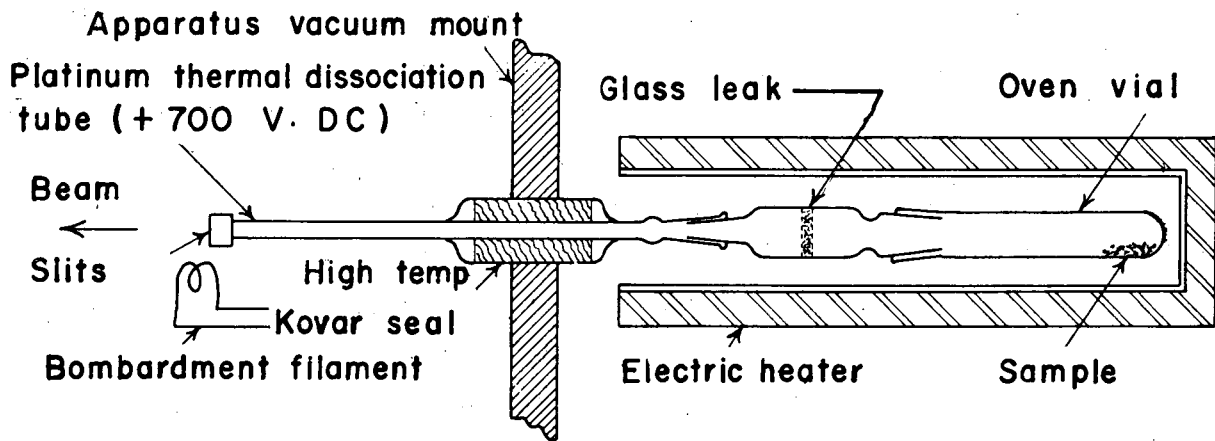
The astatine-211 was produced by an  $\alpha, 2n$  reaction on a bismuth target, prepared by melting bismuth metal onto an aluminum supporting foil.<sup>54,55</sup> The energy of the alpha particles was held below 29 Mev to limit production of the highly active and dangerous astatine-210. The astatine was separated from the target by an evaporation technique proposed by Barton, Ghiorso, and Perlman.<sup>56</sup> The target is heated to 700° C in air within a stainless steel crucible which is shown in Fig. 22. The top of the crucible is closed by a water-cooled cylinder, to which is clamped a platinum disc upon which the astatine is collected. It is necessary to mix the astatine with a carrier substance and iodine was chosen because of the similarity of its chemical properties to those of astatine. The platinum foil is placed in an evacuated flask with approximately 200 milligrams of iodine and heated by an induction heater to drive off the astatine; an intimate mixing of the astatine and iodine is ensured by distilling the mixture several times from one end of the vial to the other. An atomic beam of astatine is produced by thermal dissociation of the At-I complex in a platinum tube heated by electron bombardment at its snout to approximately 700° C. The dissociator is shown in Fig. 23. In order to prevent absorption of the active material on the walls of the glass containing vessel and connecting tube the glass must be maintained at a temperature of at least 100° C. At this temperature iodine has an appreciable vapor pressure and the flow of mixture to the platinum tube is controlled by a slow leak manufactured by partially fusing a fritted glass chemical filter before its insertion into the flow line. With this arrangement a 70-80% dissociated beam of astatine is obtained.



MU-18037

Fig. 22. Evaporator used to separate astatine from bismuth.





MU-15447

Fig. 23. Thermal dissociator used to produce a beam of astatine atoms.

### 3. Bismuth

Axensten, Johansson, and Lindgren have measured the nuclear spins and hyperfine structures of a number of bismuth isotopes (masses 203 through 206). They produced  $\text{Bi}^{203}$  (12hr) and  $\text{Bi}^{204}$  (12 hr) by the reaction  $\text{Pb}(p, kn) \text{Bi}$  using 35-Mev protons, and  $\text{Bi}^{205}$  (15day) and  $\text{Bi}^{206}$  (6.3day), by the reaction  $\text{Pb}(p, kn) \text{Bi}$ , with 25-Mev deuterons. The bismuth is separated from the lead by the following procedure: the lead target, together with some natural bismuth (0.3 to 3 mg depending upon the strength and half life of the sample) is dissolved in 6 N nitric acid and the solution neutralized with ammonia. A few drops of dilute nitric acid are added until the solution becomes slightly acid and the white precipitate of bismuthyl nitrate is redissolved. By the method of internal electrolysis (described below) the bismuth is deposited on nickel wires wound into spirals. The spirals are put directly in the oven. If electrolysis is carried out at a temperature just below the boiling point of the solution and at the proper acidity, the procedure is very rapid and a yield of about 80% is obtained in about 20 minutes. In earlier experiments nickel powder was used instead of nickel wires but the powder exhibited the strange property of producing beams whose intensity was almost independent of the oven temperature. This phenomenon was perhaps due to difficulties experienced by the nickel atoms in diffusing through the powder. The oven in these experiments was built from molybdenum, the front consisting of a channeled snout heated by electron bombardment to a temperature of  $1400^{\circ} \text{C}$ ; at this temperature bismuth molecules dissociate to atoms. The main body of the oven is heated by conduction along the snout to  $650^{\circ} \text{C}$ , when the vapor pressure of bismuth is adequate to maintain a beam.

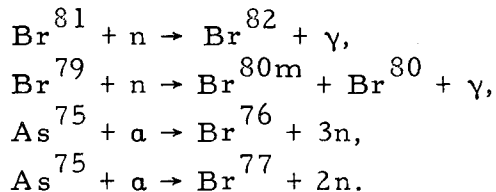
Marino<sup>59</sup>, working with 6.4day  $\text{Bi}^{206}$ , produced this isotope by bombarding .015 in. thick water-cooled lead foils with 24-Mev deuterons. The dominant reaction is  $\text{Pb}^{206}(d, 2n) \text{Bi}^{206}$ . The method of internal electrolysis was used to separate the bismuth from the lead. The target, with 5 mg of bismuth, is dissolved in 60 ml of nitric acid (20% dilute) and the solution heated gently and neutralized with concentrated ammonium hydroxide solution. Three ml of concentrated nitric acid are added to the neutral solution and the volume increased to 250 cc by adding water. The lead anodes of the electrolysis tank are surrounded with dialyzing tubing and the two compartments so

formed are filled with a 5% solution of lead nitrate acidified with nitric acid. The solution containing the dissolved target is heated to 85° C, poured into the main volume of the tank, and a platinum wire electrode is inserted. When the electrode and the two lead anodes are connected together the bismuth plates out upon the wire which is placed directly into the oven with the dissociation snout, shown in Fig. 9d. The plating operation takes about an hour.

Alpert<sup>40</sup> has completed measurements on  $\text{Bi}^{210}$  (5day). Here 3-gm cylinders of bismuth metal were irradiated for several weeks in a pile. The beams were produced simply by heating the cylinders in a dissociation-oven (Fig. 9d).

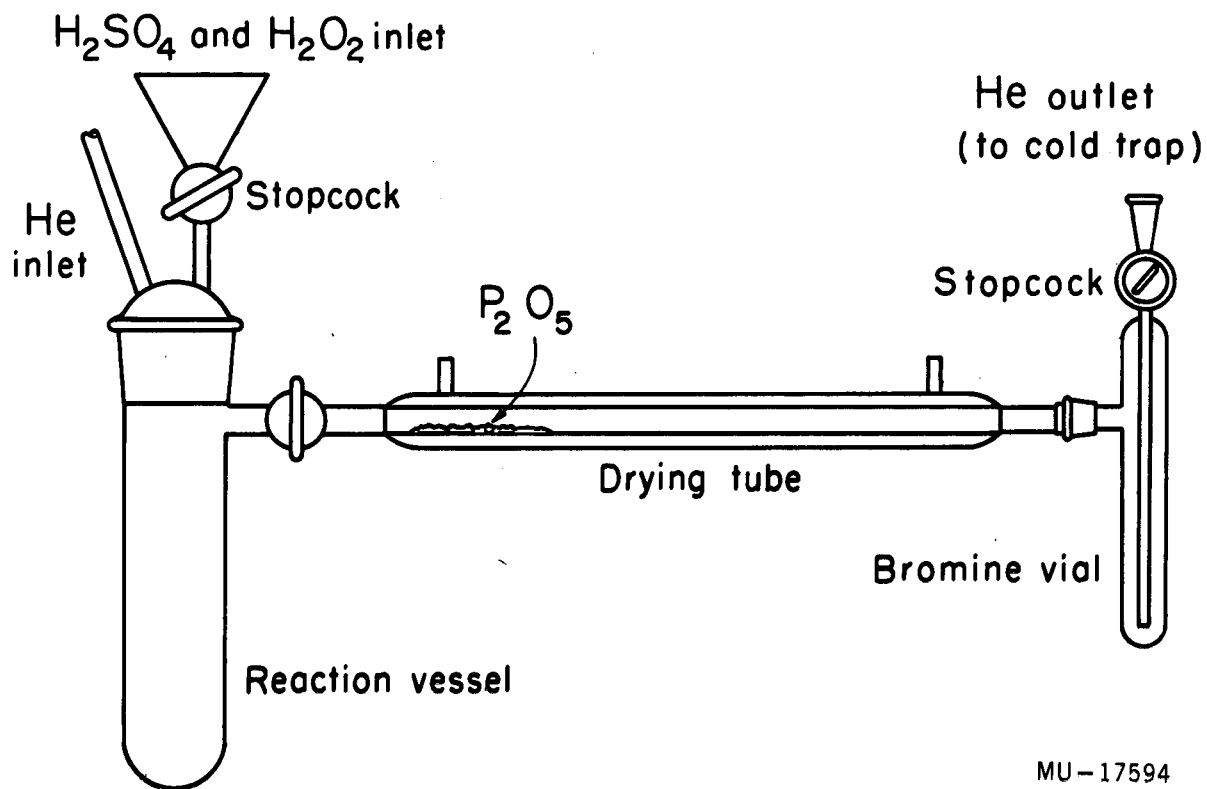
#### 4. Bromine

The isotopes Br<sup>76</sup> (17hr), Br<sup>77</sup> (57hr), Br<sup>80m</sup> (4.5hr), Br<sup>80</sup> (18 min), and Br<sup>82</sup> (35hr) have been investigated by Green.<sup>61, 28</sup> Bromine-82, -80m, and -80 were produced by neutron bombardment of anhydrous KBr powder while Br<sup>76</sup> and Br<sup>77</sup> were manufactured by bombarding arsenic powder with alpha particles. The relevant reactions are:



Arsenic powder in a "cats eye" holder (Sec. IV-B) was bombarded with 30 $\mu$ A of alpha particles at an energy of 46 Mev to produce Br<sup>76</sup>. The alpha particle energy was reduced by 8 Mev when Br<sup>77</sup> was manufactured in order to reduce contamination by unwanted Br<sup>76</sup>.

A beam of bromine atoms was produced by dissociating bromine vapor in a discharge tube;<sup>28, 41</sup> the tube is shown in Fig. 12. In order to obtain free bromine from the KBr powder and arsenic the apparatus shown in Fig. 24 was employed. Natural bromine carrier in the form of a weighed amount of KBr powder is placed in the reaction vessel together with the active material and dry helium passed through the apparatus to remove moist air. A small flow of helium is maintained until the chemistry is complete. The right hand vial is cooled in liquid nitrogen and a few cc. of sulphuric acid slowly added to the left hand vessel sufficient to cover the sample; small amounts of hydrogen bromide, bromine, and sulphur dioxide, are liberated at this stage. The bromine is released in a controlled way by adding hydrogen peroxide drop by drop and is carried by the helium flow into the cold vial where it condenses. A considerable quantity of SO<sub>2</sub> condenses with the bromine and this is removed by allowing the vial to stand open in ice water for a few minutes; after sealing with a glass stopper the vial is cooled in liquid nitrogen and transported to the apparatus.



MU-17594

Fig. 24. Apparatus used to obtain bromine from KBr powder.

The flow of bromine to the discharge tube is controlled by a leak. Two types of leak were successfully used, the first made by packing glass powder into a 3-mm diameter glass tube to a length of about 1/8 in. and heating with a soft flame until the powder appears to adhere to the outer tube. Though a useful leak of this type can only be made by trial and error, once a leak of approximately the correct size has been found the bromine flow can be controlled by adjusting the amount of carrier material introduced during the chemistry, and the temperature of the bromine vial during operation; when these leaks clog they can easily be cleaned by pumping on one end and sparking into the other with a Tesla coil.

A second class of leaks, constructed in a manner suggested by Gordon<sup>62</sup>, was also used. These leaks can be manufactured with a reproducible leak rate, and once an operating leak rate has been decided upon, any number of more or less identical leaks can be made. For bromine at room temperature a leak with an apparent diameter (as defined by Gordon) of 2.3 microns had an effusion rate of 0.3 cc/hr and operated very successfully.

The bromine atoms were collected upon brass or stainless steel buttons coated with freshly evaporated silver. The buttons had to be stored under vacuum until they were used to protect the silver surface.

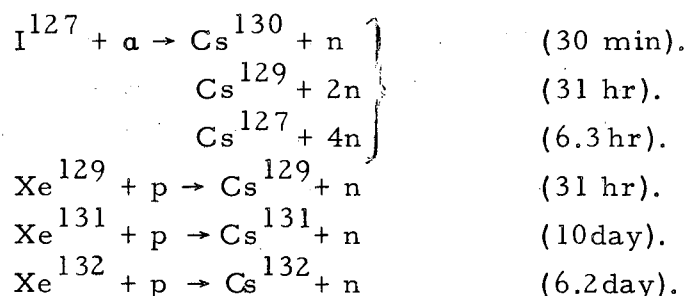
## 5. Cesium

The cesium isotopes, probably because of the ease with which they can be produced and detected, have been extensively studied in atomic beam experiments.

The long-lived neutron-excess isotopes have been the monopoly of the M. I. T. group who have been the main users of channeled ovens to conserve activity.<sup>63, 64</sup> They detected the radioactive atoms by surface ionization subsequently counting the ions with an Allen-type photomultiplier tube; the radioactive atoms were differentiated from each other and from their stable cesium carrier with a mass spectrometer. Two channeled ovens that have been used for cesium (and other alkali metals) are shown in Figs. 15, 16, and 17; these ovens are described in detail in Sec. V-C.

Davis<sup>63</sup> in measurements on Cs<sup>137</sup> operated with 210  $\mu$ C of Cs<sup>137</sup> (30y) diluted to 1 part per thousand by Cs<sup>133</sup>. The Cs<sup>137</sup>, available as the chloride, was placed in an oven with excess sodium azide (NaN<sub>3</sub>) and heated to 300° C to decompose the azide; the liberated sodium displaces cesium from the CsCl. Stroke et al. in experiments on Cs<sup>134</sup> (2.3yr), Cs<sup>135</sup> (3 $\times$ 10<sup>6</sup> yr) and Cs<sup>137</sup> (30yr), using samples in the form of CsCl in HCl neutralized the acid with Na<sub>2</sub>CO<sub>3</sub> and liberated cesium by heating the residue with chips of freshly cut potassium metal in the oven shown in Fig. 17. It is of interest to note that the latter authors were able to measure the hyperfine anomalies between Cs<sup>133, 134, 135, 137</sup> with one 150 mc sample of Cs<sup>134</sup> and a sample containing only 15 mc of Cs<sup>135, 137</sup> and that after surface ionization detection and analysis with a mass spectrometer, resonance counting rates observed with the Allen tube were about 50/sec.

The radioactive neutron-deficient isotopes have been investigated by Shugart and Nierenberg.<sup>65, 66</sup> They were produced by bombarding iodine in the form of anhydrous BaI<sub>2</sub> powder with  $\alpha$  particles, and xenon gas with protons. The important reactions are:



The bombardments of  $BaI_2$  powder were carried out using the target holder shown in Fig. 3a, and the following chemistry employed to obtain the active cesium in the form of cesium iodide. The barium iodide (100 to 200 milligrams) is dissolved in 200 cc of water containing known amounts of cesium iodide (carrier (5 to 50 milligrams) and excess of ammonium carbonate added to precipitate the barium as barium carbonate, leaving ammonium iodide and cesium iodide in solution. The solution is filtered and the ammonium iodide and the cesium iodide concentrated by boiling the solution to dryness. Upon further heating, the dry ammonium iodide sublimes and the cesium iodide remains on the container walls. The low sublimation temperature of ammonium iodide ( $551^{\circ}C$ ) enables the separation to be made very readily. After sublimation, the radioactive cesium iodide together with the stable carrier cesium iodide is dissolved in approximately 0.2 cc of distilled water and transferred to a small metal cup which fits into the atomic beam oven. The solution must be evaporated to dryness and, in order to avoid boiling or sputtering, the cup is placed on an aluminum plate heated by a steam bath, and slow stream of air is passed across the liquid surface. An infrared lamp may also be used reduce the evaporation time.

Figure 6 shows the container used for the production of cesium isotopes by bombardment of xenon gas with protons; the external dimensions are  $19 \times 5 \times 1\frac{1}{2}$  inch. The container is made of cast aluminum and is cooled by water circulating through copper tubing welded to the walls. The window assembly contains two 1-mil thick aluminum foils to separate the xenon gas from the cyclotron vacuum: the proton beam passes from the cyclotron vacuum through the first foil into an air space maintained at a pressure of  $1/2$  an atmosphere. After traversing this air space (approximately  $1/2$  inch) the protons pass through the other 1-mil foil to enter the region containing xenon at atmospheric pressure.

The chemistry required to obtain the cesium in a useful form is simplicity itself. The xenon is removed by attaching the container to an auxiliary bulb cooled to liquid nitrogen temperature: all the cesium activity remains in the target vessel and can be recovered by washing it with 100 to 200 cc of water containing a few milligrams of cesium iodide carrier and a trace of hydroiodic acid. Three washings remove 90% of the activity.



The main body of water is then boiled away and the latter steps of the foregoing procedure entered upon.

Beams of cesium are finally produced by adding sodium-free calcium to the cup containing cesium iodide, and heating.

Cohen<sup>67</sup>, in one of the first atomic-beam experiments performed on a radioactive isotope, measured the spin and hyperfine structure of cesium-134 m (3.1hr). The sample, which consisted of 100 mg of neutron-irradiated cesium chloride (CsCl), was heated in a monel metal oven with metallic barium chips; at 450° C a strong beam of cesium is produced. Goodman and Wexler in a later measurement on the same isotope irradiated natural cesium in quartz vials in a flux of  $1.5 \times 10^{13}$  neutrons/cm<sup>2</sup>/sec. The vials were crushed in an oven in a dry box attached to the apparatus. The oven of cold rolled steel was supported on three legs one of which was made of tungsten and another of tantalum. The two legs and their function with the oven constituted a thermocouple which allowed the oven temperature to be measured between 100 and 1500° C.

## 6. Copper

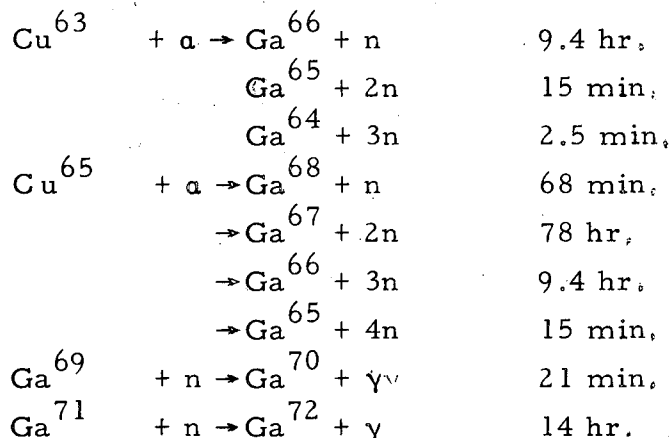
Dodsworth has produced beams of the radioactive copper isotopes  $\text{Cu}^{61}$  (3.3hr) and  $\text{Cu}^{62}$  (10min). The  $\text{Cu}^{61}$  is produced by bombarding foils of cobalt metal with 34 Mev alpha particles through the reaction  $\text{Co}^{59}(\alpha, 2n)\text{Cu}^{61}$ . The foils are dissolved in 16N nitric acid and the solution boiled to dryness. The residue is dissolved in 6N HCl, diluted to 3N with water, and  $\text{H}_2\text{S}$  gas is bubbled through the solution to precipitate the copper as copper sulphide. The latter is dissolved in dilute nitric acid and the solution boiled to drive off the  $\text{H}_2\text{S}$ . The copper is now electroplated upon a fine platinum wire where it forms a mush-like deposit which is scraped off into a warm tantalum oven, (warm to evaporate moisture).

Copper-62 is a daughter of  $\text{Zn}^{62}$  (9hr). The latter isotope is produced by the reaction  $\text{Ni}^{60}(\alpha, 2n)\text{Zn}^{62}$  in nickel foil. Some  $\text{Cu}^{61}$  is produced simultaneously, presumably by the reaction  $\text{Ni}^{58}(\alpha, p)\text{Cu}^{61}$ , and the  $\text{Cu}^{61}$  produced plays an important role in the experiment. The nickel foil is dissolved in aqua regia together with a few milligrams of zinc and copper, boiled to dryness, and the steps of the chemistry described above repeated, with the following modifications.

The first precipitate of  $\text{CuS}$  is dissolved in dilute  $\text{HNO}_3$ , diluted and set aside; it contains the  $\text{Cu}^{61}$  and  $\text{Cu}^{62}$  but the latter rapidly decays. An hour is allowed to elapse to reestablish secular, equilibrium between  $\text{Zn}^{62}$  and  $\text{Cu}^{62}$  and a measured amount (~1cc) of  $\text{Cu}^{61}$  solution is added to the main body of solution containing  $\text{Cu}^{62}$ . A small sample of this solution is taken for decay to establish the initial ratio of  $\text{Cu}^{62}$  to  $\text{Cu}^{61}$  atoms, and the final steps of the chemistry entered upon and completed as rapidly as possible. The whole oven load of  $\text{Cu}^{61}$  and  $\text{Cu}^{62}$  is evaporated onto the collecting button as rapidly as possible (5 min) and the button decayed. At resonance the ratio of  $\text{Cu}^{62}$  to  $\text{Cu}^{61}$  is enhanced, and when it is extrapolated to zero time it can be compared with the ratio obtained from the earlier sample. Enhancement ratios at resonance of 20:1 have been observed.

7. Gallium

Hubbs,<sup>70</sup> Worcester<sup>37</sup> and Ehlers<sup>71</sup> have produced gallium isotopes by alpha-particle bombardment of copper foil, and neutron bombardment of gallium metal. The following reactions predominate:



The cyclotron targets, cut from commercial sheet copper .010 in. thick, were water cooled on the back surface and bombarded in the internal beam of a cyclotron at currents ranging from 40 to 90  $\mu\text{A}$  for times from 0.5 to 10 hrs. Some targets immediately after bombardment registered as much as 1000  $\gamma/\text{hr}$  at a foot and the rather elaborate chemistry necessary to separate the gallium from copper was carried out by remote control in a lead-walled "cave". All attempts to produce beams of gallium by heating the target foil in an oven directly, failed, and the procedure to be described for preparing the oven load (a modification of a method described by Swift in 1924) was the only successful one found. The copper target is dissolved in 15 cc of 10 N  $\text{HNO}_3$  containing 20 to 40 mg of gallium carrier. The solution is boiled to drive off most of the liquid and 60 cc of 6N  $\text{HCl}$  saturated with diethyl ether is added. The solution is placed in an extraction flask with 50 cc of ether saturated with  $\text{HCl}$  and stirred vigorously with a shaded pole arcless stirring motor to avoid the possibility of an ether explosion. The gallium chloride dissolves in the ether which is washed twice more with 25 cc of 6N  $\text{HCl}$  to remove all traces of copper; this last step appears to be essential to avoid difficulties later. The gallium chloride is extracted from the ether into 15 cc of water and ion ten N sodium hydroxide added until the pH is 5; bromcresol-green indicator turns blue at this pH value. At this point the pH is measured on a meter and a 10% solution of acetic acid added drop by drop until the pH is exactly 5.5. The gallium, which

precipitates as  $\text{Ga}(\text{OH})_3$ ; is separated with a centrifuge and redissolved in one or two drops of 10N  $\text{NaOH}$ . The gallium metal is then electroplated onto a short length of 0.006-in. diam. platinum wire with a current of 0.5 amps at 8 v and the globule of gallium which forms at the tip of the wire is scraped off into the oven. A complete separation takes about 45 min exclusive of the electroplating operation. The short-lived  $\text{Ga}^{68}$  is electroplated for 15 min, the long lived  $\text{Ga}^{67}$  for 45 min; the separation efficiencies in these two cases are about 60 and 85% respectively. Beams could only be produced from ovens of carbon operating at  $1300^\circ\text{C}$ ; these carbon ovens are described in Sec. V-A. Ovens of iron and tantalum are useless as the gallium alloys immediately with these materials, an effect noted by Renzetti<sup>73</sup> in his work as the stable gallium isotopes.

8. Gold

The gold isotopes that have been studied in atomic beam experiments are listed below:

<u>Isotopes</u>	<u>Half-Life</u>
Au <sup>191</sup>	3 hr
Au <sup>192</sup>	4.8 hr
Au <sup>193</sup>	17.5 hr
Au <sup>194</sup>	34 hr
Au <sup>195</sup>	180 day
Au <sup>196</sup>	5.6 day
Au <sup>197</sup>	stable
Au <sup>198</sup>	2.7 day
Au <sup>199</sup>	3.2 day

Ewbank<sup>74, 75</sup> produced gold-191, -192, -193 by bombarding iridium foils with alpha particles, and gold-194, -195, -196 by bombarding platinum foils with protons and deuterons. Stable iridium possesses two stable isotopes: Ir<sup>191</sup> (38.5%) and Ir<sup>193</sup> (61.5%) and though all gold isotopes from Au<sup>191</sup> to Au<sup>196</sup> can be produced by alpha-particle bombardment of iridium alone, certain difficulties encountered with iridium targets make alternative means of production more acceptable. Iridium is impervious to strong acids and bases and cannot be dissolved easily; its melting point is high (2454°C) and this makes it difficult to distill the gold from it in an evaporator. The bombarded iridium foil was placed directly into the oven and heated. Gold does not distill at a uniform rate from iridium and the beam falls with time.

The platinum targets were bombarded with protons and deuterons using the internal beam of a cyclotron, and though it has since been found that stable gold beams can be effused from platinum contained in closed carbon crucible inside a tantalum oven, the gold at the time these experiments were performed was separated chemically from the platinum. The platinum is dissolved in hot concentrated aqua regia (a process taking several hours) together with some gold carrier, and the solution evaporated to dryness by gentle heating to avoid the reduction of gold chloride to gold. The residue is dissolved in 6 N HCl and an equal volume of ethyl acetate added. The solution separates into two parts with the organic layer containing the gold and this is separated off and washed with HCl. The ethyl acetate is evaporated off, the gold chloride

dissolved in water and the metallic gold precipitated by blowing  $\text{SO}_2$  across the surface of the solution. If the  $\text{SO}_2$  is blown through the solution very small gold crystals are formed, and the losses encountered while handling these crystals can be great. Recently Ewbank and Chan have found that very stable beams of gold are obtained when the gold is evaporated from a completely enclosed carbon liner contained within a tantalum oven. The liner has a small snout which projects through the oven wall, and the exit slit is cut into the end of this snout.

Gold-198 and 199 have been studied by Christiansen et al.<sup>76</sup> The gold-198 is prepared by irradiating natural gold in a pile. The atomic beam oven was made of molybdenum metal and heated to  $1150^\circ\text{C}$ . Gold-199 is produced by the (np) reaction in platinum in a pile. The platinum (about 0.5 grams) was irradiated for about a week and contained about 9 millicuries of activity.

9. Helium

Commings and Kusch<sup>77</sup> have performed a Stern-Gerlach-type experiment designed to set an upper limit to the magnetic moment of  $\text{He}^6$  (0.83 sec). The  $\text{He}^6$  is produced by neutron bombardment of  $\text{Be}(\text{OH})_2$  in a pile; the gas-handling system is described in the literature.<sup>78</sup> The beam, after passing through the deflecting magnets, enters a long, narrow detector channel which terminates in a cylindrical aluminum cavity. The dimensions of the channel and cavity are such that the probability for re-emergence of an  $\text{He}^6$  atom during the decay half-life is small. The  $\beta$ -rays emitted during the decay penetrate the thin walls of the cavity (0.25 mm thick) and enter a plastic scintillation crystal surrounding the cavity. The scintillator is connected to a photomultiplier tube by a light pipe.

10. Indium

Marino<sup>79</sup> has made a study of the indium isotopes,  $\text{In}^{109}$  (4.3 hr),  $\text{In}^{110\text{m}}$  (5 hr), and  $\text{In}^{111}$  (2.8 day) and has measured their nuclear spins and hyperfine interaction constants. The isotopes are manufactured by bombarding 0.005-in. silver foils with 48-Mev alpha particles in a cyclotron; the useful reactions are of the type  $\text{Ag}(\text{d}, \text{kn}) \text{In}$ . Alpha particle beam currents of 90  $\mu\text{A}$  are employed and bombarding times range from 3 to 10 hours, the latter exposure being used for the 2.8d isotope. To prepare the oven load, the central portion of the bombarded foil is dissolved in a minimum amount of concentrated nitric acid together with 25 mg of indium carrier. The solution is evaporated to dryness and 10 ml of distilled water added and the silver precipitated from solution as the chloride by adding concentrated hydrochloric acid. After the chloride is separated in a centrifuge, concentrated ammonium hydroxide is added drop by drop to the solution, and the precipitate of indium hydroxide again separated in a centrifuge. The hydroxide is dissolved in concentrated HCl added drop by drop and when solution is complete, two drops of formic acid are added and the indium electroplated onto a platinum wire using a current density of 3 amps/cm<sup>2</sup>. The indium is scraped off the wire into a carbon oven. The separation time, using the above procedure, is about 3 hours and the yield 80%. Indium beams can be produced from either tantalum or carbon ovens.

Goodman and Wexler<sup>80</sup> produced  $\text{In}^{114\text{m}}$  (50 day) and  $\text{In}^{116\text{m}}$  (54 min) by irradiating 0.005-in. foils of indium metal in a pile. In the case of  $\text{In}^{114\text{m}}$  the foils were irradiated for six months and a graphite oven used to form the beam. For  $\text{In}^{116\text{m}}$  a graphite oven was loaded with 25 mg of indium foil, and the oven itself placed in a pile for 8 hours; the irradiated oven could be placed immediately into the atomic-beam apparatus.



## 11. Iodine

Garvin<sup>55</sup> and others have made an extensive series of measurements on the radioactive iodine isotopes.<sup>81, 82, 83, 84</sup> The isotopes are produced in many different ways (see below) but, with the exception of  $I^{128}$  (25 min), the final stage of chemistry terminates with a sample of about 200 mg of stable iodine carrier in which is mixed the active isotope. In order to produce a beam of iodine atoms, it is necessary to dissociate iodine molecules. This has been done by two methods. The first employs the radiofrequency discharge shown in Fig. 12 and described in Sec. V-A. The iodine, contained in a flask, is simply attached to the discharge tube; the vapor pressure of iodine at room temperature is adequate for the maintenance of a discharge. The tube is an efficient dissociator of iodine (and bromine) molecules and estimates indicate that the emergent beam is 80 to 90% atomic. The second method makes use of the thermal dissociator (made of platinum) shown in Fig. 23 but used without the heating jacket or leak. When the snout of the dissociation tube is heated to 700° C a beam of iodine is produced that is at least 60% dissociated.

$I^{123}$  (13 hr) and  $I^{124}$  (1r day) are produced by bombarding powdered antimony metal with 48-Mev alpha particles in the holder shown in Fig. 4. Several attempts were made to separate the iodine from the antimony by distilling the iodine out of the metal under vacuum and collecting it upon a cold surface, but yields were only moderate (~50%) and difficulties arose as the antimony tended to volatilize with the iodine. A simple and efficient chemical procedure was devised which resulted in the recovery of 80-90% of the iodine. The antimony powder is dissolved in concentrated HCl (a process which can be speeded up by adding a little hydrogen peroxide to the solution) and a few milligrams of sodium iodide carrier added. When the solution is adjusted slightly past the neutral point with NaOH, the antimony precipitates as antimony oxychloride which can be filtered off. The precipitate is washed with a few ml of NaOH containing NaI, and the iodine precipitated from filtrate by the addition of  $NaNO_2$  in dilute sulfuric acid. The iodine is extracted into carbon disulfide and the solution added to a flask containing approximately 200 milligrams of elemental iodine; the mixture is well shaken and the carbon disulfide evaporated off under vacuum.

$I^{126}$  (13 day) and  $I^{130}$  (12.6 hr) are made by bombarding tellurium metal with protons through the  $Te(p,n)I$  reaction. The tellurium is melted onto a 0.025 in.-thick aluminum foil and bombarded in the holder shown in Fig. 4.

After bombardment the tellurium is dissolved in nitric acid and the solution made basic with sodium hydroxide. If formic acid is now added, the tellurium precipitates as tellurium oxide and the iodine can be extracted from the solution by acidifying with  $H_2SO_4$  and adding  $NaNO_2$  as above. The iodine can then be extracted into carbon disulphide containing stable carrier iodine. A faster but less efficient method uses an evaporation method similar to that employed for astatine 211. The evaporator is shown in Fig. 22. When the tellurium is heated to  $700^\circ C$ , the iodine is released and collected upon the cooled platinum disc which is subsequently washed in NaOH solution when the iodine dissolves.

$I^{132}$  (2.3 hr) (is obtained as a daughter of  $Te^{122}$  (77 hr). The  $Te^{122}$  is obtainable from Brookhaven National Laboratory in a special container which can be flushed with ammonium hydroxide solution to remove the iodine. The generator regains its secular equilibrium in 12 hours and fresh samples of  $I^{132}$  can be removed at 12-hour intervals if desired.

$I^{131}$  (8 day) and  $I^{133}$  (21hr) are obtainable commercially as sodium iodide. The iodine is easily obtained in free form by the above method ( $NaNO_2 + H_2SO_4$ ).

$I^{135}$  (6.7 hr) is produced (together with other iodine isotopes) as a fission product when  $U^{238}$  is bombarded with 24-Mev deuterons. After bombardment, the uranium is dissolved in HCl. The solution is diluted with water and  $NaNO_2$  added to bring down free iodine. The iodine is efficiently separated from other fission products when it is extracted into carbon disulphide.

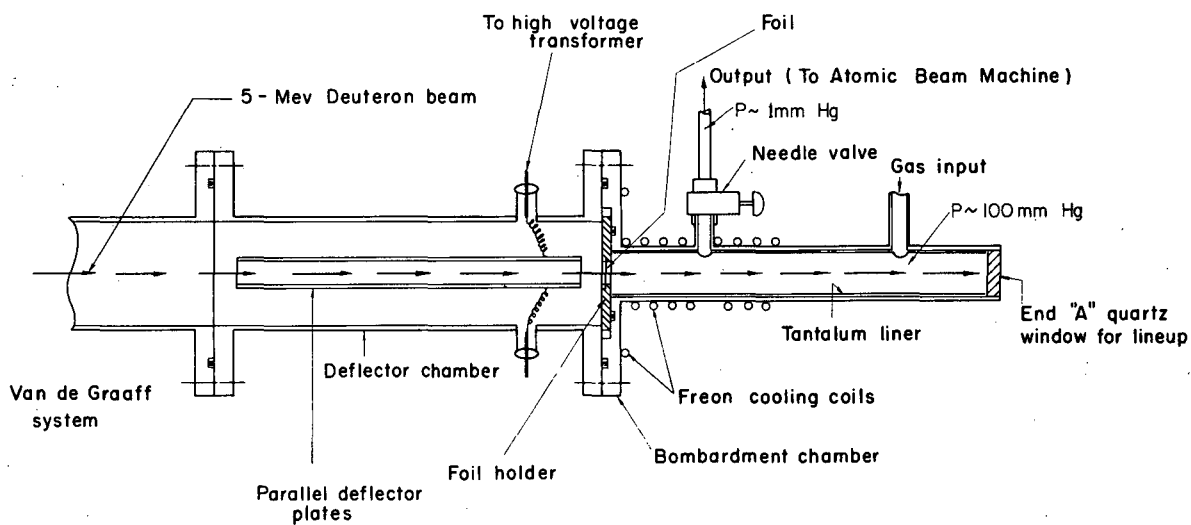
Sherwood and Ovenshine<sup>84</sup> have produced beams of  $I^{128}$  (25 min) and  $I^{130}$  (12.6 hr) using a solid-gold dissociation oven maintained at  $750^\circ C$ . They produced  $I^{128}$  and  $I^{130}$  by neutron bombardment of stable iodine and maintained their iodine sample at ice temperature outside the vacuum system.

## 12. Oxygen

Commins\* and Feldman at Columbia have recently observed resonances in a beam of  $O^{15}$  (121sec). The  $O^{15}$  is produced by bombarding nitrogen gas with 5-Mev deuterons; the reaction is  $N^{14}(d, n)O^{15}$  and proceeds with a cross section of 100 mb. A schematic diagram of the source assembly is shown in Fig. 25. The deuterons ( $7 \times 10^{-6}$  amp) enter a nickel bombardment chamber containing nitrogen gas at a pressure of 100 mm Hg, mixed with small quantities of NO and  $O_2$ . Before entering the chamber the beam passes between two plates 20 cms long and 3/16 in apart between which is applied 2500 volts at 60 cps; the beam, in this way, is swept back and forth over the entrance foil to minimize chances of burnout; the foil itself is a sheet of molybdenum 0.00025 in.-thick covered with a thin layer of evaporated platinum to prevent attack by reaction products produced in the gas. The bombardment chamber is lined with tantalum sheet to minimize the neutron background at the atomic beams apparatus that arises from neutrons produced by "stripping" when deuterons collide with the chamber walls. Tantalum has a small stripping cross section for deuterons.

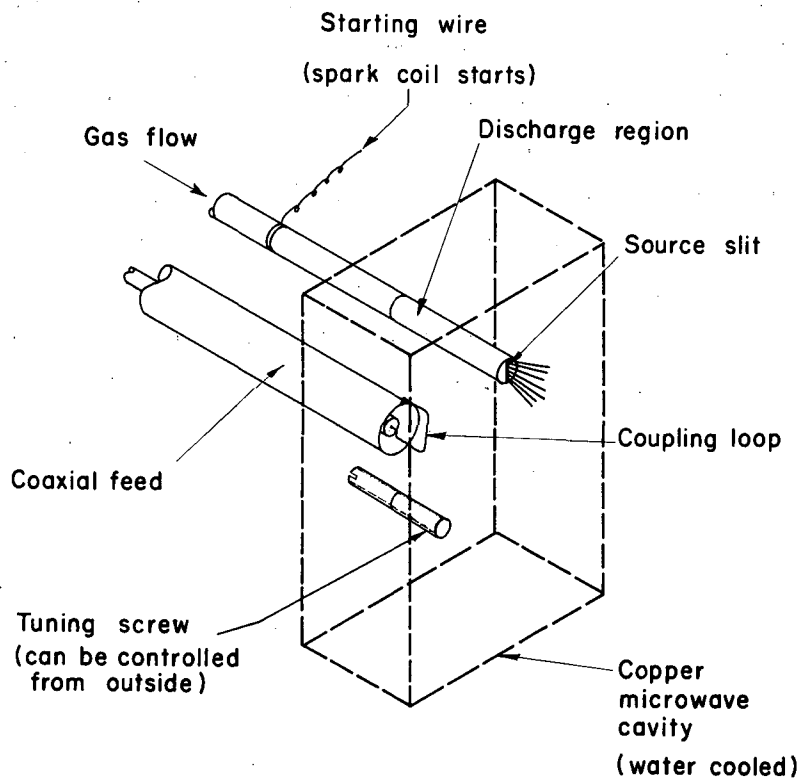
When the deuteron beam passes through the gas, it produces  $O^{15}$  by nuclear reaction and at the same time numbers of nitrogen and oxygen atoms by dissociation of the  $N_2$ , NO, and  $O_2$ . The  $O^{15}$ , if it is not lost to the walls, combines with a free nitrogen or oxygen atom to form  $NO^{15}$  or  $O^{16}$ , and it is this molecule, carried by the gas flow through the needle valve, that is transported to the atomic beams apparatus 30 feet away. If pure nitrogen is used without NO or  $O_2$  additive, the yield of  $O^{15}$  is reduced by a factor of three. The gas containing  $NO^{15}$  and  $O^{15} O^{16}$  is dissociated in the rf discharge tube shown in Fig. 26. The tube is quartz, 7/32 in.-od and 5/32 in.-id, and a slit 0.002 in. wide is ground into the closed end. The last 1-1/2 in. of the tube passes through a microwave cavity and is arranged to lie on a voltage antinode within the cavity. The cavity is excited at 2460 Mc/sec with a magnetron oscillator (Raytheon RK 5609) which is coupled into the cavity by an adjustable loop. The discharge is started with a spark coil. The operating pressure in the discharge tube is about 1 mm Hg; the needle valve in the flow line reduces the pressure in the line from 100 mm Hg in the bombardment chamber to approximately 1 mm Hg in the line. The flow conditions are such that about half the 0.15 atoms produced in the chamber survive to reach the discharge tube.

\* E. D. Commins, private communication.



MU-21844

Fig. 25. Source for the production of  $O^{15}$ .



MU-21846

Fig. 26. Discharge tube for dissociation of molecules containing  $O^{15}$ .

### 13. Potassium

Zacharias,<sup>86</sup> in a now famous paper, describes the first atomic-beam "flop in" resonance experiment, performed on naturally occurring  $K^{40}$  and, incidentally, the first such experiment on a radioactive isotope, for  $K^{40}$  is unstable against  $\beta$ -decay with an half-life of  $1.3 \times 10^9$  yr.<sup>87</sup>

Petersen et al.<sup>25</sup> have produced  $K^{43}$  (22 hr) by bombarding argon with alpha-particles in a container similar to that shown in Fig. 6 and described in Sec. V-A. The reaction is of the type  $A^{40}(\alpha p) K^{43}$  and proceeds with a fairly large cross section because of the low  $Z$  of the target nucleus (see Sec. II-D). The potassium is recovered by washing the target container with distilled water containing about 30 mg of potassium chloride carrier. Three washings with 200 ml of water are adequate to remove most of the activity. The solution is reduced in volume by boiling and transferred to the oven with a pipette. After evaporation to dryness, finely divided calcium metal is added and the oven is ready for use. Potassium is released when the oven is heated to about  $400^\circ$  C.

14. Rare Earths

Cabezas<sup>39</sup> had performed atomic-beam experiments on neutron-activated rare earth isotopes listed below.

<u>Isotope</u>	<u>Half-Life</u>
Ce <sup>143</sup>	33 hr
Pr <sup>142</sup>	19.1 hr
Nd <sup>147</sup>	11.6 day
Sm <sup>153</sup>	47.0 hr
Gd <sup>159</sup>	18.0 hr
Tb <sup>160</sup>	172.0 day
Ho <sup>166</sup>	27.2 hr
Er <sup>169</sup>	9.4 day
Er <sup>171</sup>	17.5 hr
Tm <sup>170</sup>	129.0 day
Tm <sup>171</sup>	1.9 yr

In all cases metal pellets of high purity contained inside of a glass or quartz capsule, were bombarded with neutrons. The air inside the capsule was removed by a stream of rare gas before sealing, to prevent oxidation of the metal. Beams of all these elements were produced simply by heating the pellets in a sharp-lipped tantalum crucible contained inside a tantalum oven. The oven and liner were identical to that shown in Fig. 11. Of all these metals only praesodymium showed any tendency to creep and this difficulty was overcome by working with samples of high specific activity at as low a temperature as possible,

White<sup>88</sup> has produced beams of Lu<sup>176m</sup> (3.7 hr) in an exactly similar way but has found that a creep can be minimized by putting some tantalum carbide powder into the oven with the pellet.

## 15. Rubidium

The rubidium isotopes,  $\text{Rb}^{81}$  through  $\text{Rb}^{84}$ , are produced most easily by bombarding bromine with alpha particles, though the reaction  $\text{Br}(\alpha, \text{kn})\text{Rb}$ .  $\text{Rb}^{81}$  (4.7hr) is of historical interest: Hobson<sup>89</sup> and Hubbs<sup>90</sup> performed the first zero moment atomic-beam experiment on a radioactive isotope using  $\text{Rb}^{81}$ . Sunderland<sup>91</sup> later studied this isotope using a resonance apparatus.

The target material in the form of anhydrous barium bromide powder is bombarded in the holder shown in Fig. 3a. The rubidium is separated from the target material using the following procedure. The barium bromide is dissolved in 5 to 10 ml of water containing 1 to 15 mg of rubidium bromide carrier, and the barium is precipitated as the carbonate by addition of a large excess of ammonium carbonate. The remaining liquid is boiled to dryness and the residue heated to  $550^{\circ}\text{C}$ . The ammonium bromide sublimates away to leave the rubidium bromide which is dissolved in a few drops of water, placed into an oven cup, and dried. A beam of rubidium is formed when the rubidium bromide is heated, with calcium chips in a stainless steel oven to between  $400^{\circ}$  and  $600^{\circ}\text{C}$ .

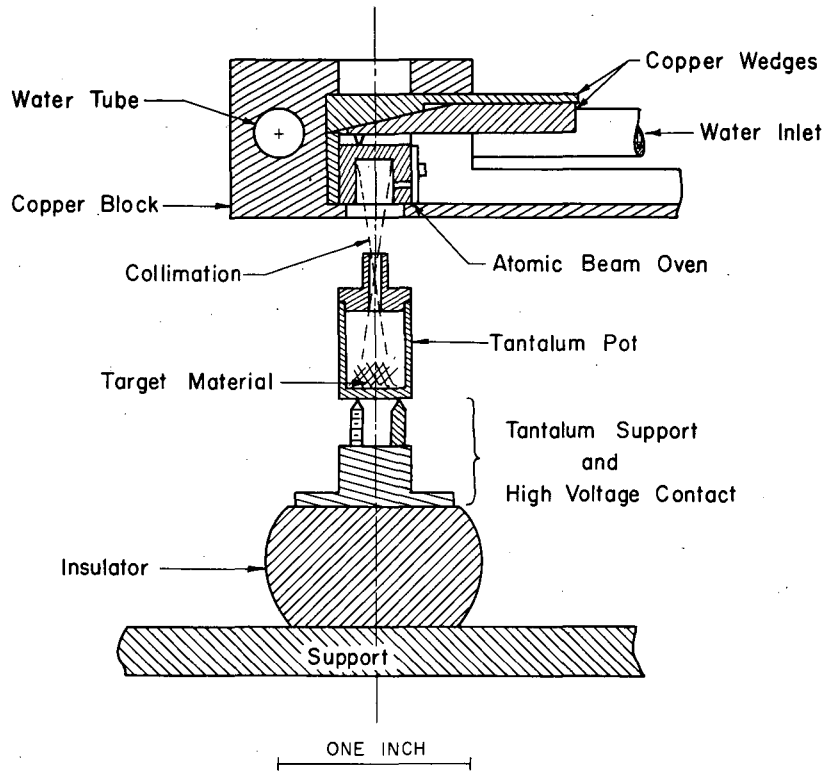
$\text{Rb}^{84}$  can also be produced more efficiently by the  $(\text{pn})$  reaction on krypton gas,  $\text{Kr}^{84}(\text{pn})\text{Rb}^{84}$ . Two liters of krypton gas at atmospheric pressure are bombarded, in the container shown in Fig. 6, with 12 Mev protons. After allowing short-lived activities to decay, the krypton is condensed in another container attached to the first and the activity is washed out with several hundred ml of water containing Rb Br carrier. The main body of water is boiled away and the concentrated solution transferred to an iron oven cup which is thoroughly dried. Calcium is added and the beam produced as before.



## 16. Silver

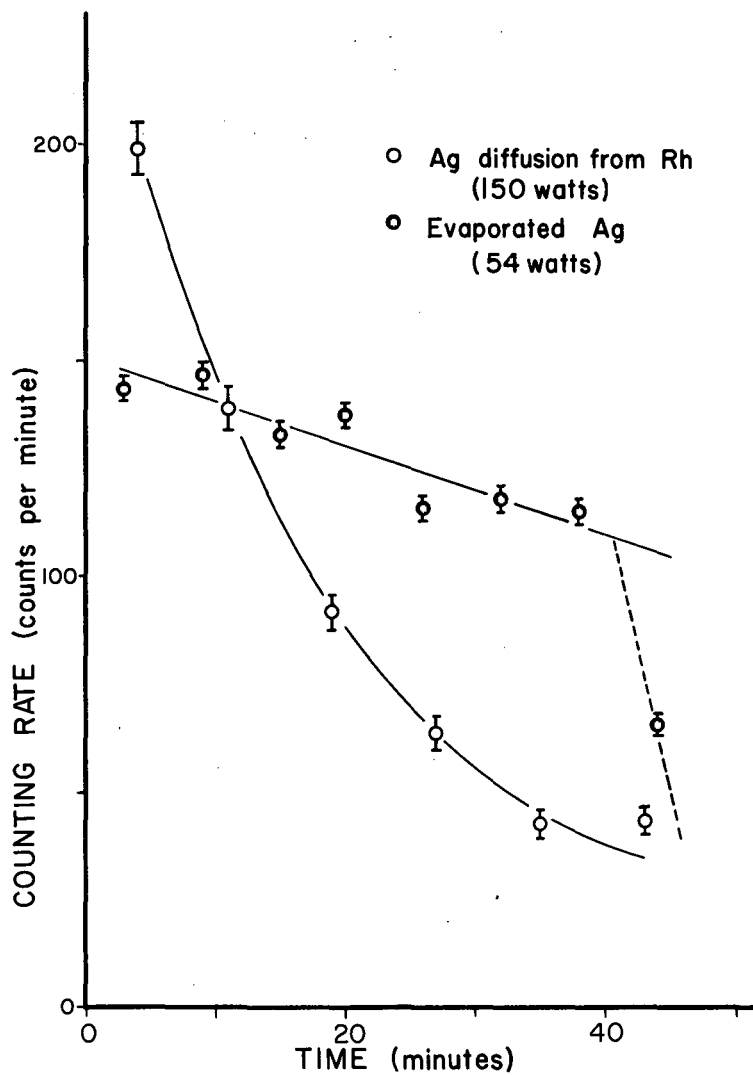
Ewbank has produced the short-lived silver isotopes  $\text{Ag}^{103}$  (1.1 hr),  $\text{Ag}^{104}$  (27 min),  $\text{Ag}^{104}$  (1.2 hr), and  $\text{Ag}^{106}$  (24 min) for atomic-beam experiments by bombarding foils of rhodium metal with 48 Mev alpha-particles; the silver is produced by the reaction  $\text{Rh}(\alpha, \text{kn}) \text{Ag}$ .<sup>74, 92, 93</sup> He has also produced  $\text{Ag}^{105}$  (40 day) and  $\text{Ag}^{105}$  (8.3 day) by bombarding palladium foils with 12 Mev protons, while Reynolds et al.<sup>94</sup> by use of 18-Mev protons on palladium, have made experiments with the same two isotopes. Both workers have produced beams by simply heating the target foils in ovens of tantalum and molybdenum, but Ewbank finds it more satisfactory to separate the silver from the target, particularly in the case of rhodium. In order to obtain a useful beam of silver from rhodium, the rhodium must be heated close to its melting point; but if the rhodium is allowed to melt, the silver is lost in a very short time. It is difficult to obtain the precise control of oven temperature necessary for the maintenance of a stable silver beam; at constant temperature the beam decays rapidly with time. In Fig. 27 is shown the evaporator used by Ewbank<sup>74</sup> to distill silver from rhodium directly into the tantalum oven cup, and Fig. 28 exhibits the gain in beam stability that comes about through use of the evaporator.

In order to determine the optimum foil thickness and alpha-particle energy for production of the short-lived isotopes, a stacked-foil experiment was performed. Twelve small pieces of 0.001 in. Rh foil was placed in a target holder so that an alpha-beam passed through each foil in turn. The assembly was exposed to an alpha-particle beam (15  $\mu\text{A}$  48 Mev) for one second. The foils were counted in rotation for several hours in three counters; (a) an x-ray crystal counter set to count the Ag K-capture x-ray, (b) a counter set to count the higher energy  $\gamma$  radiation, and (c) a continuous flow gas ionization chamber. The decay curve for each foil was analyzed into two components, one of 25 min half-life, the other of 70 min half-life. The results<sup>74</sup>, shown in Fig. 29, provide two important pieces of information: (1) the alpha-particle energy required to produce a maximum amount of each isotope, and (2) the most efficient method of counting each isotope. It will be seen that 1.1 hr  $\text{Ag}^{103}$  and 1.2 hr  $\text{Ag}^{104}$  decay principally by K-capture while 27 min  $\text{Ag}^{104}$  and 24 min  $\text{Ag}^{106}$  decay by positron emission or isomeric transitions. The most advantageous combination of rhodium target foils and aluminum degrading foils and aluminum degrading foils used by Ewbank are summarized in Table II.



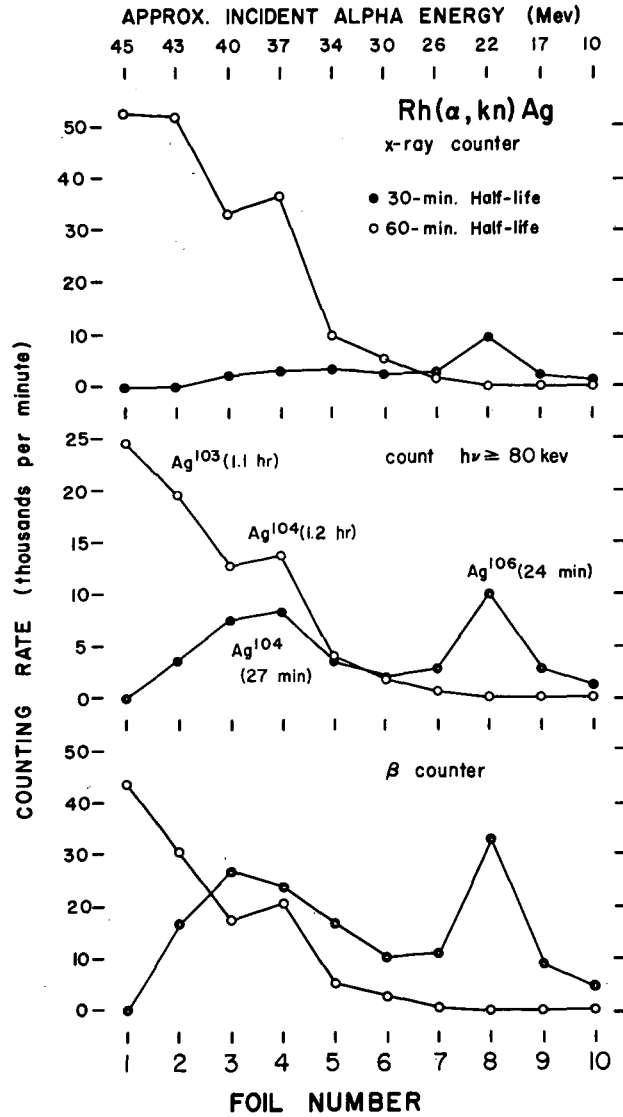
MU-17483

Fig. 27. Evaporator to distill silver from rhodium into oven cup.



MU-16791

Fig. 28. Comparison of effusion rates of silver from rhodium, and evaporated silver from a tantalum oven.



MU-17482

Fig. 29. Results of a stacked foil experiment to determine the optimum alpha energies for the production of certain silver isotopes.

Table II

Optimum thickness of target foils for production of silver isotopes		
Isotope	Thickness of Al Degrading foils (mils)	Thickness of Rhodium target (mils)
Ag <sup>103</sup>	≤ 1	2
Ag <sup>104</sup>	5 to 7	4
Ag <sup>106</sup>	~ 21	2

The control required of the oven temperature when silver beams are produced by heating irradiated palladium foils is even more critical than in the case of rhodium, but silver can be easily separated chemically from palladium as follows: The palladium is dissolved in aqua regia with silver carrier; the silver precipitates as silver chloride while the palladium is dissolving. The precipitate is washed, dissolved in ammonium hydroxide, and reprecipitated by boiling off the ammonia. The washing and purification is continued until the AgCl is white and free from the orange-colored PdCl<sub>2</sub>. The purified AgCl is dissolved once more in NH<sub>4</sub>OH, and ammonium iodide is added to precipitate silver iodide. The iodide is washed, dried, and loaded into the oven and when heated gradually, the silver iodide decomposes into silver and iodine. Further heating produces a beam of silver.

## 17. Sodium

Sodium is of particular interest historically. Davis<sup>95,96</sup> in his study of the long lived isotope  $\text{Na}^{22}$  (2.58 yr) was the first to employ a channeled oven as a means of conserving active material, while Bellamy<sup>97</sup> and Smith<sup>98</sup> with short-lived  $\text{Na}^{24}$  (14.9 h) performed the first atomic-beam magnetic resonance experiment on a short-lived radioisotope.

Davis produced  $\text{Na}^{22}$  by the (d,  $\alpha$ ) reaction on magnesium in a cyclotron;  $\text{Mg}^{24}(\text{d}, \alpha)\text{Na}^{22}$ . The sodium was separated chemically from the magnesium using a chemical procedure described by Irvine and Clark<sup>99</sup>, and converted to sodium azide ( $\text{NaN}_3$ ). A sample of azide containing  $230\mu\text{C}$  of  $\text{Na}^{22}$  diluted to  $1:10^4$  with  $\text{Na}^{23}$  was put into a channeled monel metal oven (see Fig. 15 and Sec. V-C) and heated to  $300^\circ\text{C}$  to yield sodium and free nitrogen. The beam was detected upon on hot tungsten filament, and the ions counted with an Allen-type photomultiplier tube; the evaporated sodium ions were analyzed by a mass spectrometer. It is of interest to note that during one 16-hr run only  $4 \times 10^{-22}$  moles of  $\text{Na}^{22}$  was evaporated from the oven and that the beam intensity during this time amounted to only a few thousand atoms per second. Radio-frequency resonances were observed with less than 100 atoms/sec striking the photomultiplier detector. Bellamy and Smith in their work on  $\text{Na}^{24}$  irradiated a one gram sample of metallic sodium in a neutron flux of  $5 \times 10^{10}$  neutrons/cm<sup>2</sup>/sec for a week; the ratio of  $\text{Na}^{24}$  to  $\text{Na}^{23}$  was  $2 \times 10^{-8}$  corresponding to a specific activity of 200 mc/gram. The oven was made of monel metal and similar to the one employed by Davis. The experiment of Bellamy and Smith is further noteworthy in that there for the first time in a beam resonance experiment counters were used to detect the resonated atoms. The beam was allowed to fall upon on hot oxidized tungsten filament; the sodium ions produced were attracted by an electric field to a brass collector plate and the activity absorbed on the plate subsequently counted.

## 18. Thallium

Thallium isotopes have been the subject of atomic-beam experiments by Brink<sup>100, 101</sup>, Marino<sup>59</sup>, and Lindgren and Johannson.<sup>102</sup> Brink produced  $Tl^{197}$  (2.7 hr),  $Tl^{198m}$  (1.9hr) and  $Tl^{199}$  (7.4 hr) by bombarding gold foils with alpha particles in a cyclotron, and  $Tl^{104}$  by irradiating stable  $Tl^{203}$  in a pile. An evaporator was employed to separate the thallium from the gold; the evaporator is shown in Fig. 30. The target foil is cut into several pieces and placed with some thallium metal carrier in a steel evaporation cup; the cover of the cup has a 1/16-inch diameter hole in its center. The cup is mounted on a tungsten rod passing through a metal to glass seal in the base plate; the water-cooled brass block holds a second small tantalum oven cup directly over the hole in the top of the steel cup. A small piece of gold foil is welded to the outside of the cup containing the activated gold foil and the whole assembly heated by electron bombardment until the gold foil on the outside of the cup melts. At the temperature of molten gold the thallium diffuses to the cooled upper cup in a few seconds.

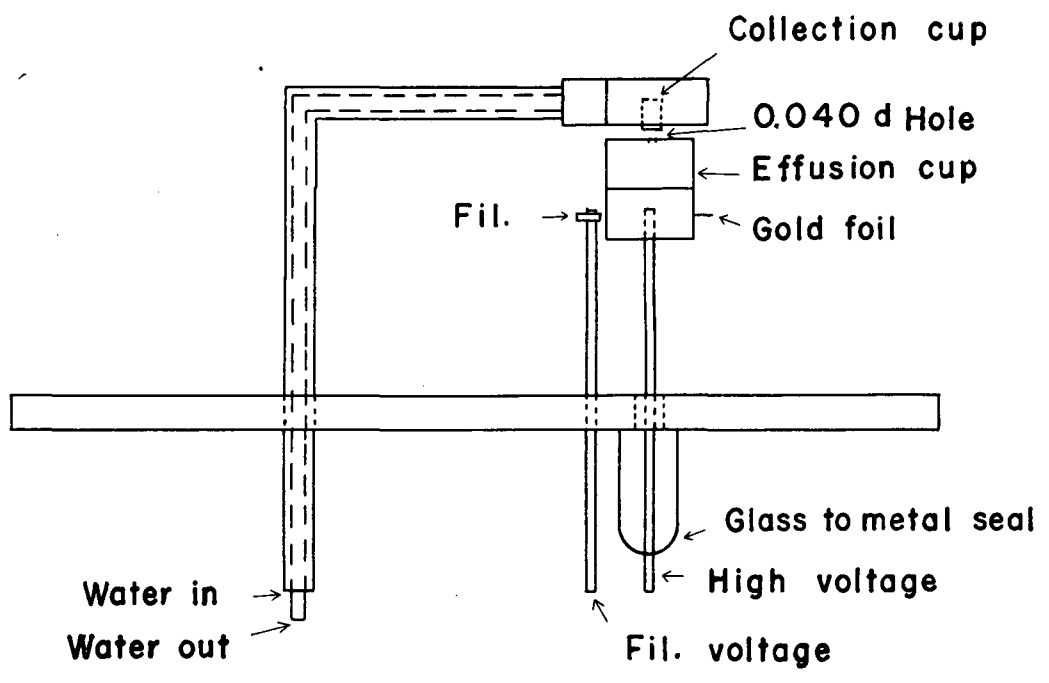
It is worth noting that the alkali metal oxides have a very low vapor pressure at 800° C, (about  $10^{-16}$  atmospheres), and their decomposition pressure is about  $10^{-5}$  atmospheres of potassium and oxygen. A few milligrams of cesium nitrate were loaded into the oven with the thallium metal and heated to a few hundred degrees C to decompose the cesium nitrate into nitrogen and cesium oxide. At a temperature between 700 and 800° C, the temperature required to produce a thallium beam, the cesium oxide itself decomposes into cesium and oxygen. The cesium beam lasts the same length of time as the thallium beam. Marino<sup>59</sup> manufactured  $Tl^{200}$  (27 hr),  $Tl^{201}$  (72 hr), and  $Tl^{202}$  (120 hr) by bombarding mercury with 24-Mev deuterons; the thallium isotopes are produced by the  $Hg(d, kn) Tl$  reactions. The mercury target was made by drilling small holes in an aluminum plate and filling them with droplets of mercury. The droplets were sealed into the plate with a Teflon sheet covered by a 1 mil aluminum foil. Approximately 60 holes were drilled into each target plate and filled with mercury.

After bombardment the target block is unloaded into a test tube by a fine tipped glass tube connected to a water aspirator. Thirty milligrams of thallium carrier are added to the mercury and part of the mixture introduced into the cup A (Fig. 31) Stopcock B is opened and stopcock C closed and a syringe

attached at D is used to draw down sufficient mercury to fill the oven E. Stopcock B is closed and C opened and the mercury forced down to fill the oven. Stopcock C is then closed and the oven heated in an helium atmosphere by the induction heater F until all the mercury has evaporated leaving behind the radioactive thallium. The process is repeated until all the mercury has been treated in this way.

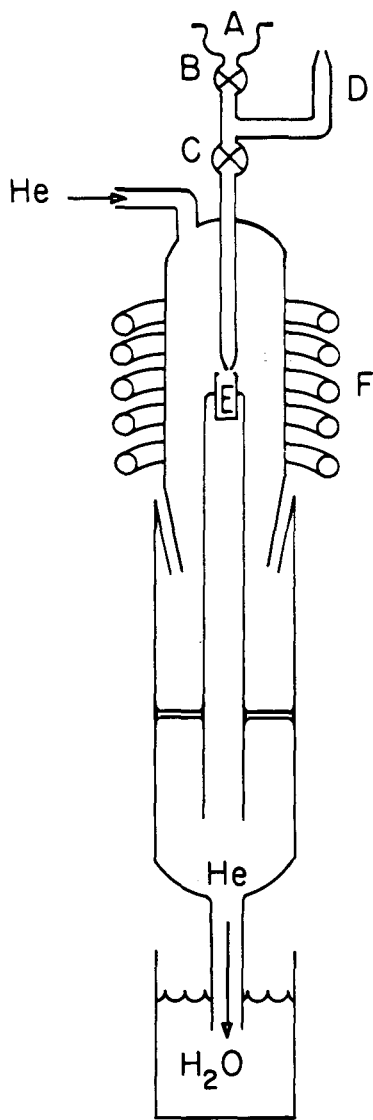
Lindgren and Johansson<sup>102</sup> bombarded stable thallium with 85-Mev protons to produce  $Tl^{198}$ . The immediate product of the bombardment is  $Pb^{198}$  which decays to the ground state of  $Te^{198}$  with a half-life of 2.3 hours. The half-life of  $Te^{198}$  is 5.3 hours. The lead is separated from the thallium by an ether extraction and electroplated, together with some milligrams of stable lead and thallium onto a copper wire which is put into the oven. This method of production of  $Te^{198}$  produces a sample, isotopically more pure than that obtained by bombarding mercury.





MU-12677

Fig. 30. Evaporator to separate thallium from gold targets.



MU-17223

Fig. 31. Apparatus used to separate thallium from mercury.  
(See text).

### 19. The Transuranium Elements

Atomic-beam experiments have been performed by Marrus and others<sup>103</sup> on selected isotopes of the artificially produced elements proto-actinium, neptunium, plutonium, americium, and curium. The experiments have established unambiguously the electronic configuration of these rare elements, and well illustrate the power of the beam method as a means of analyzing atomic structures.

Plutonium<sup>103, 38</sup>. Pu<sup>239</sup>(24,000yr). Plutonium metal is available (unhappily) in gram quantities. Great difficulty was experienced in obtaining a useful atomic beam of plutonium. The metal, when heated in a tantalum oven, alloyed and crept badly, even when contained within a sharp-lipped inner tantalum crucible. Experiments were made with refractory plutonium salts in the hope that one could be found that decomposed in the vapor phase at a pressure and temperature right for beam production, but though compounds with carbon, silicon, and oxygen were examined, none proved satisfactory. A single trial with plutonium metal in a carbon crucible indicated that the vapor pressure of the carbide is too low to be useful. Experimental ovens were constructed of molybdenum, thorium, cerium sulphide, thorium oxide, and tungsten, but only the latter metal was found to be sufficiently free from interaction with plutonium metal to be useful. Satisfactory beams were ultimately obtained at 1500° C with the tungsten containers shown in Fig. 11. The inner cup is made of tungsten metal and has a sharp lip ground the edge to control creep. In the original design, the cup was supported on a base of cerium or thorium sulphide but this was later found not to be necessary. The oven slits are thin tantalum foils spot welded to the oven surface across the exit hole. With these ovens beams of plutonium were obtained for periods as long as 15 hours at vapor pressures up to 0.5 mm Hg. The effusion rate at constant temperature is characterized by a slow monotonic drop to approximately one half the initial value, with a sudden sharp decrease over a period of less than two minutes immediately before the oven is exhausted. Though the tungsten ovens and crucibles showed no signs of deterioration after use, the tantalum slits occasionally appeared corroded.

Neptunium. 103, 104, 105 Np<sup>238</sup>(2.1day), Np<sup>239</sup>(2.35 day). Neptunium-239 is made by bombarding uranium-238 with neutrons through the reaction  $U^{238}(n, \beta) Np^{239}$ ; the reaction proceeds with a cross section of 2.76 barns. In order

to avoid excessive contamination of the target from fission products of  $U^{235}$ , a uranium target depleted of  $U^{235}$  (< 0.4%) is used, and bombardments were carried out with an integrated flux totaling approximately  $10^{19}$  neutrons/cm<sup>2</sup>.

Attempts were made at first to produce a neptunium beam by heating slivers of uranium in tantalum and tungsten ovens with inner liners similar to those used with plutonium and which are described above. However, uranium appears to creep very badly and no useful beams were obtained for periods longer than 20 minutes, when it was found that the tantalum slits had completely dissolved in the uranium metal. A very simple technique for beam production was finally developed. The uranium and neptunium are converted to the oxides by heating in air. The oxides are mixed with an excess of carbon in a tantalum container and the mixture heated to a temperature of 1700°C when a beam of neptunium atoms is formed; the heating is accompanied by considerable out-gassing, presumably due to CO formed during the reduction.

Curium,  $^{103,106}Cm^{242}$  (162 day). The experiments on curium-242 were performed with samples from a 2 cc solution containing 100µg of curium. Approximately 150 microlitres of curium solution is mixed with concentrated  $HNO_3$  and a few milligrams of uranyl nitrate added to provide a carrier. The solution is boiled to dryness and the uranyl nitrate heated to redness in air in a platinum crucible to form the oxide. In the same manner as neptunium, beams of curium are produced by heating the uranium oxide-curium oxide mixture with carbon to 1800°C in a tantalum oven.

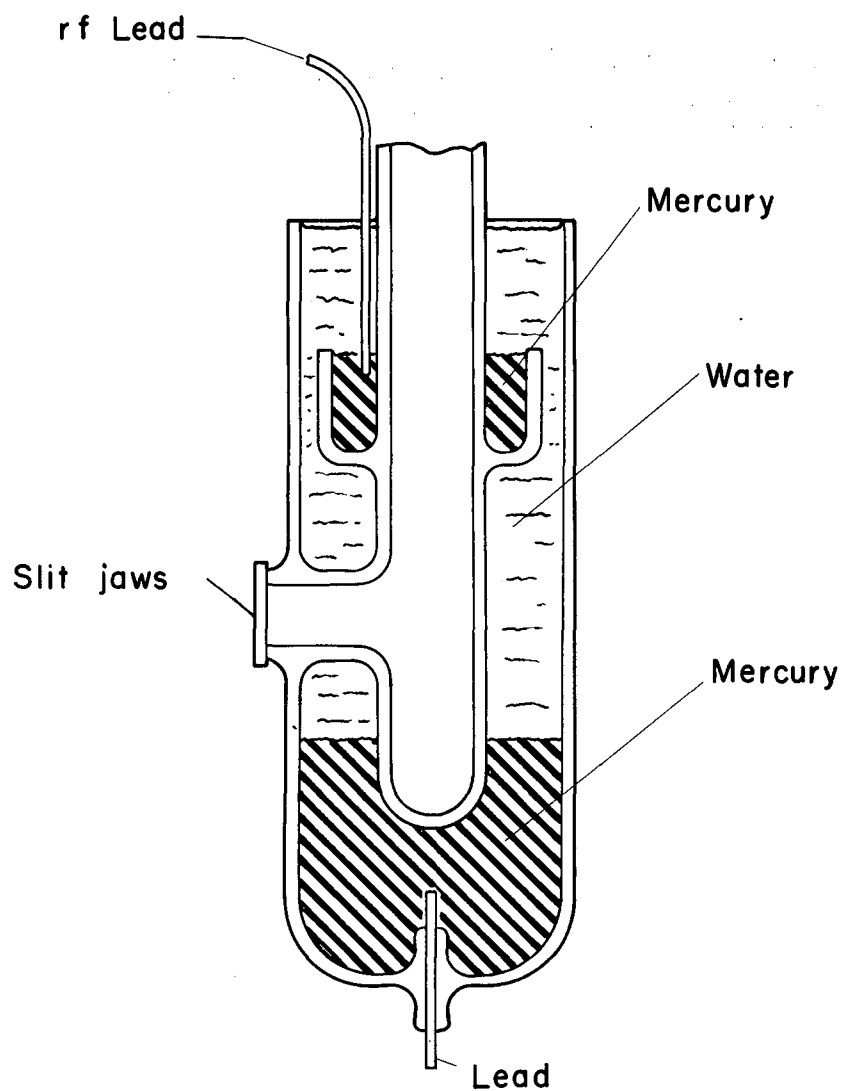
Protoactinium,  $Pa^{233}$  (27.4 day).  $Pa^{233}$  is produced by neutron irradiation of  $Th^{232}$  to produce 23.5 min  $Th^{233}$  which decays in 27.4 day  $Pa^{233}$ . Winocur<sup>33</sup> discusses the many failures that were experienced before a satisfactory beam of  $Pa$  was produced, and the final solution of the problem by an anion-exchange<sup>107</sup> separation to obtain the  $Pa$  free from thorium. The thorium target is dissolved in concentrated HCl and the solution passed through a vertical glass tube containing dowex-1 anion resin. The thorium runs through the column but the protoactinium is absorbed. The column is washed with conc. HCl to remove all traces of thorium and the protoactinium brought down with a 3 M HCl - 0.01 M HF solution, the small amount of fluoride helping to remove the protoactinium more rapidly from the resin. The solution is boiled down to a small fraction of its volume and a large excess of  $HNO_3$  added to convert the chloride to nitrate; the solution is again boiled to small volume and one drop of the concentrated solution pipetted into a tantalum oven

and evaporated to dryness; this one drop forms one oven load. An excess of carbon is added to the oven which is heated slowly to about 2500° C. At a critical temperature the beam appears suddenly and intensely and the temperature must be lowered. The effect is probably due to the sudden dissociation of protoactinium carbide.

Americium. <sup>33, 108</sup> Am<sup>241</sup> (458 yr), Am<sup>242</sup> (16 hr). Americium oxide, unlike neptunium and curium, is not well reduced by carbon and only feeble beams have been obtained using the carbon reduction technique described above. However, the oxide is well reduced by lanthanum metal, and in this way satisfactory atomic beams of americium have been formed. The procedure is as follows: A few milligrams of americium in HCl solution is converted to the insoluble Am(OH)<sub>3</sub> by the addition of concentrated (NH<sub>4</sub>) OH drop by drop. The precipitate is converted to the oxide of americium by heating it in air, in a platinum crucible, and the beam produced by heating the oxide with an excess of lanthanum metal in a sharp lipped molybdenum crucible in a molybdenum oven. At 1000° C a beam is suddenly observed; the reduction of the oxide takes place very rapidly and one must be careful to lower the temperature before too much material is lost.

20. Tritium

The hyperfine splitting in the ground state of tritium (12.5 yr) has been measured by Prodell and Kusch<sup>109</sup>, using the discharge tube shown in Fig. 32. This tube has no internal electrodes and is operated at a frequency of 4 Mc/sec. The lower end dips into a pool of mercury which acts as one of the rf terminals. The other rf connection is made through a pool of mercury held in a glass cup sealed to the discharge tube. The mercury and discharge are cooled by circulating water. The atomic tritium escapes through a slit whose glass jaws are waxed to an oven in the discharge tube.



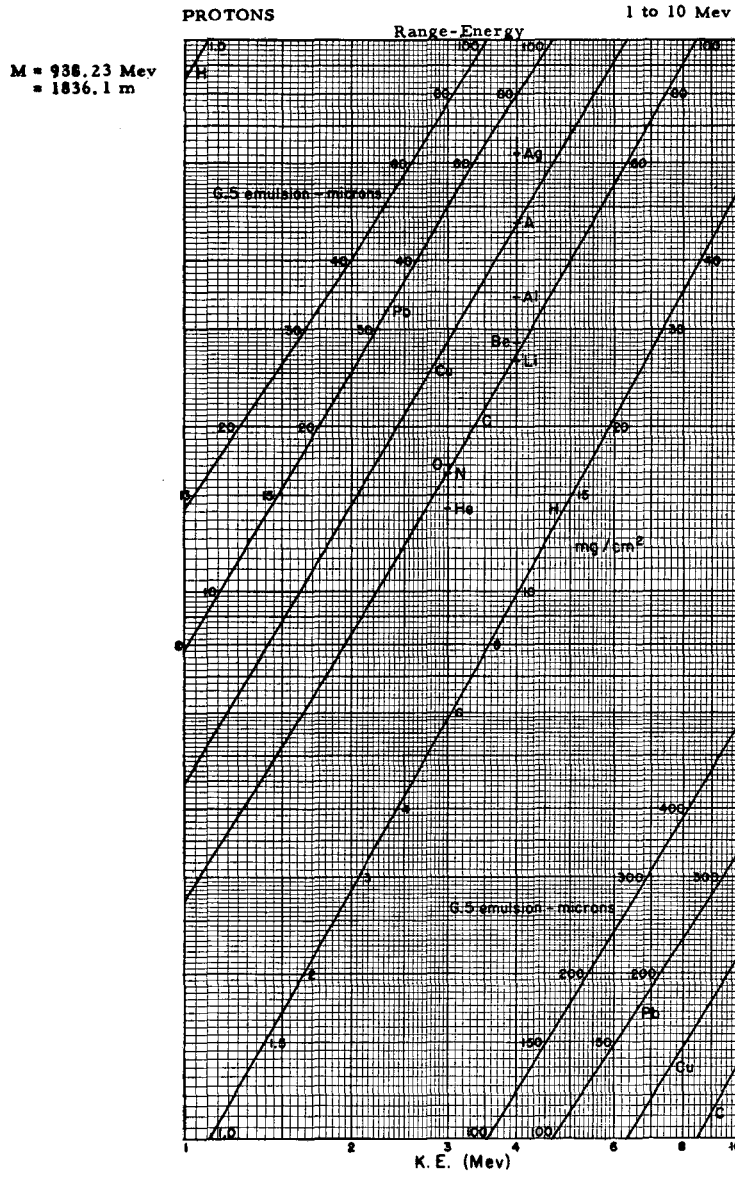
MU-21845

Fig. 32. Radio-frequency discharge tube for production of tritium atoms.

VII. APPENDIX

Figures 33 through 38. Range Energy Curves for Protons, Deuterons, and Alpha Particles in Various Materials.<sup>110</sup>





*see corrected fig  
in front of report*

Fig. 33.

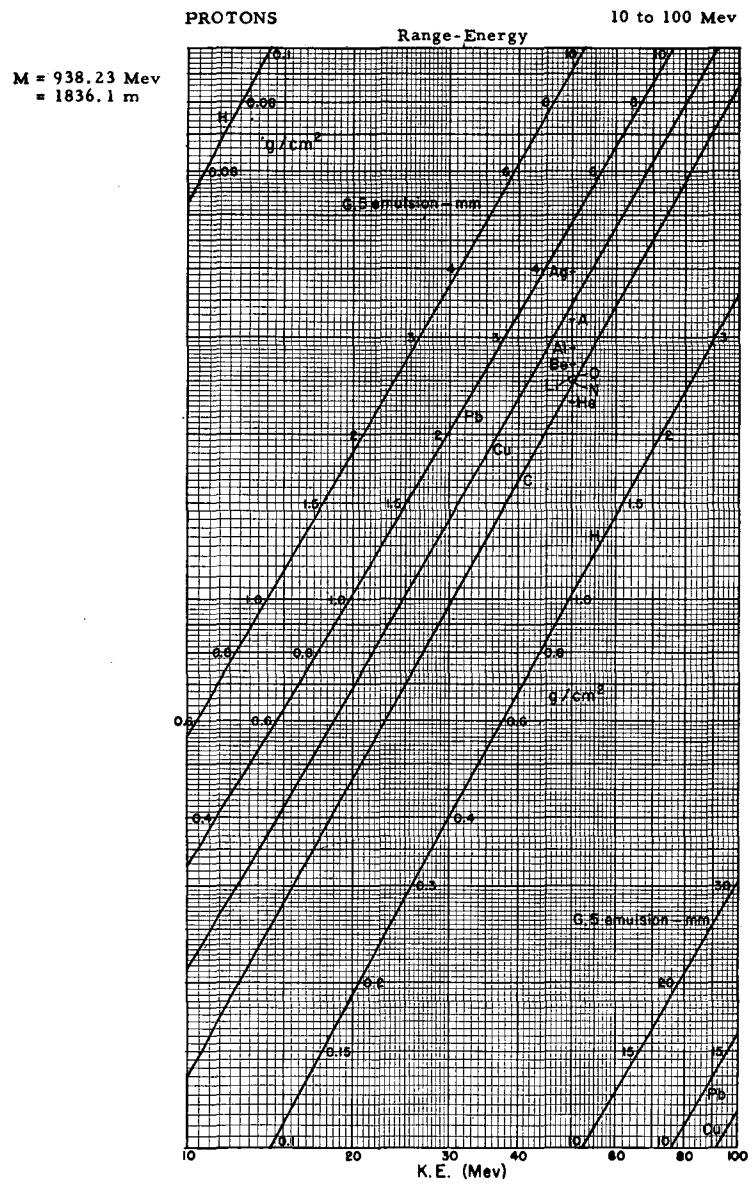


Fig. 34.

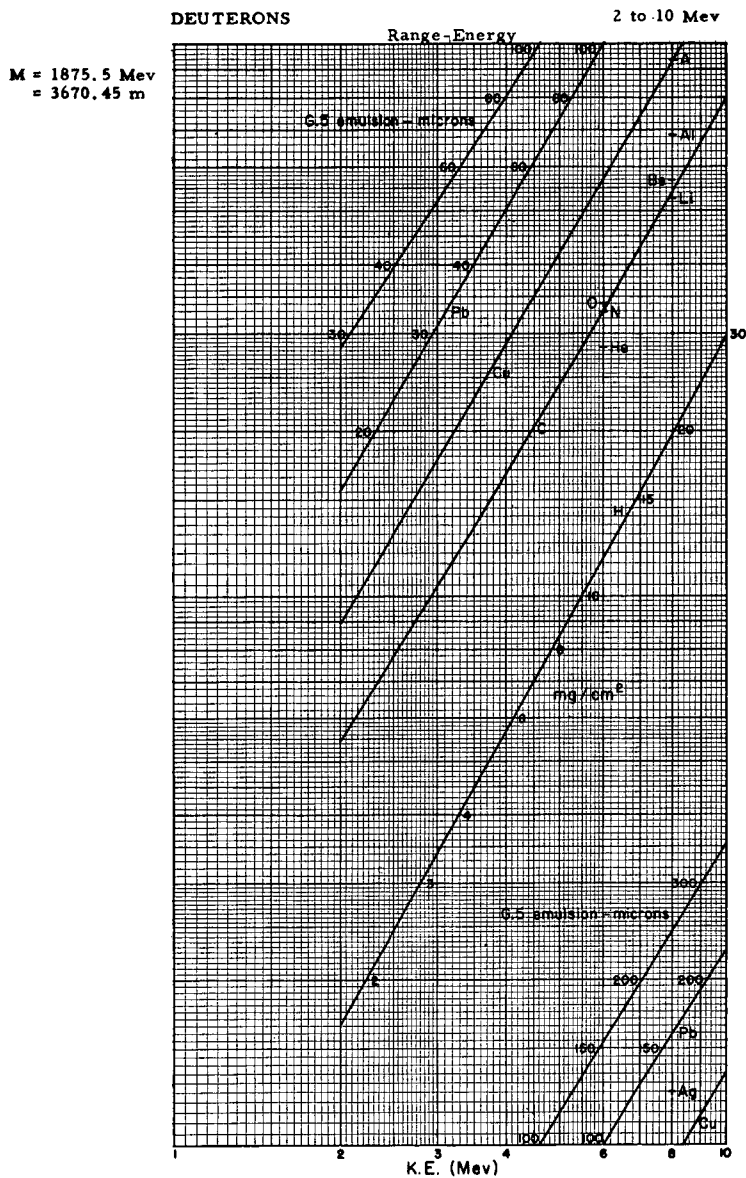


Fig. 35.

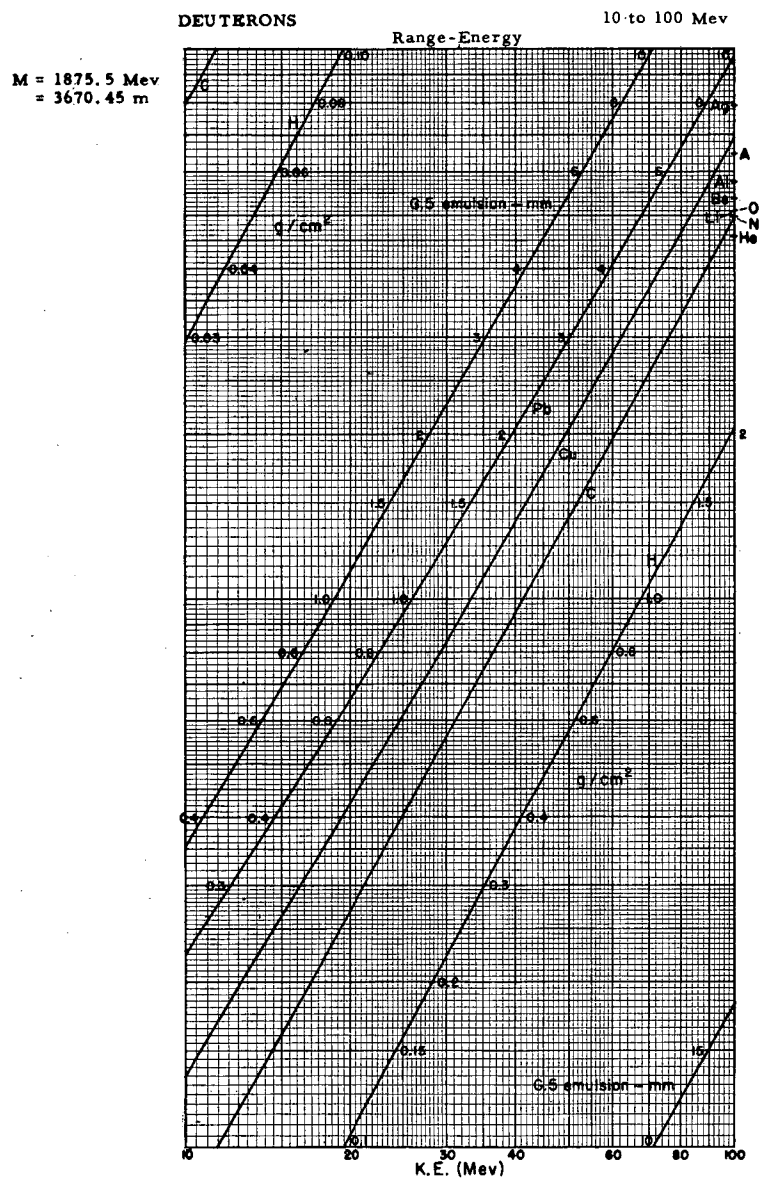


Fig. 36.

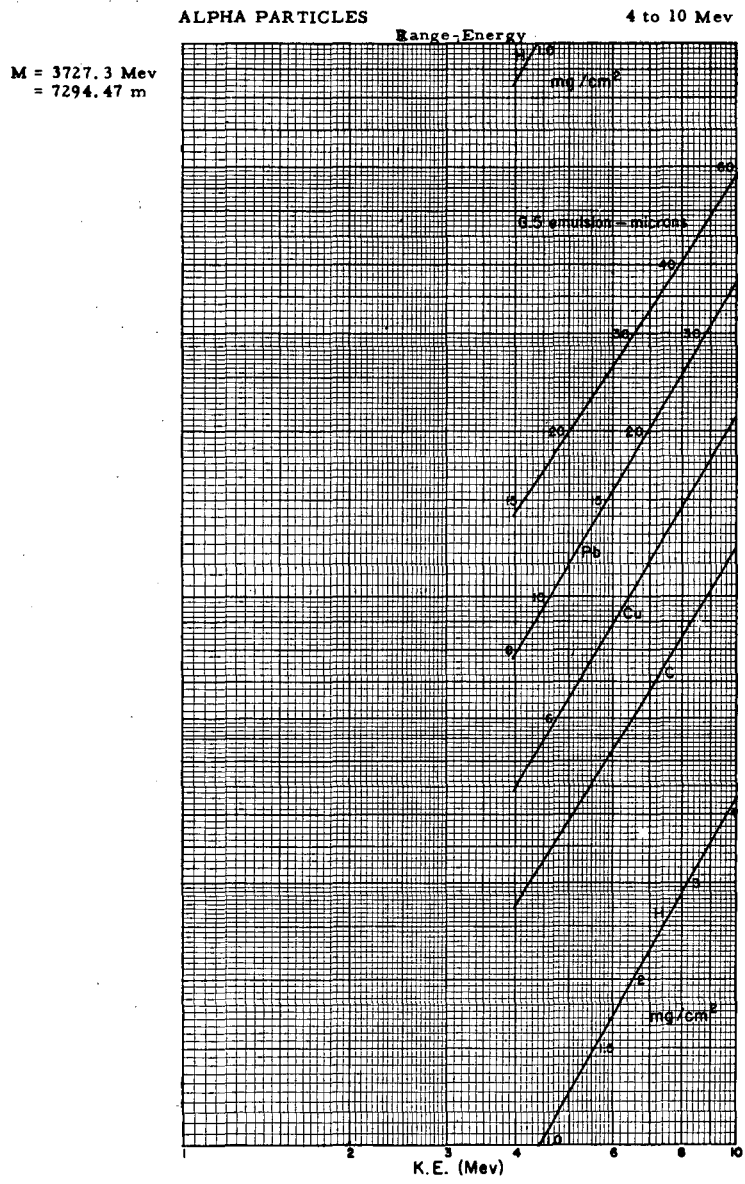


Fig. 37.

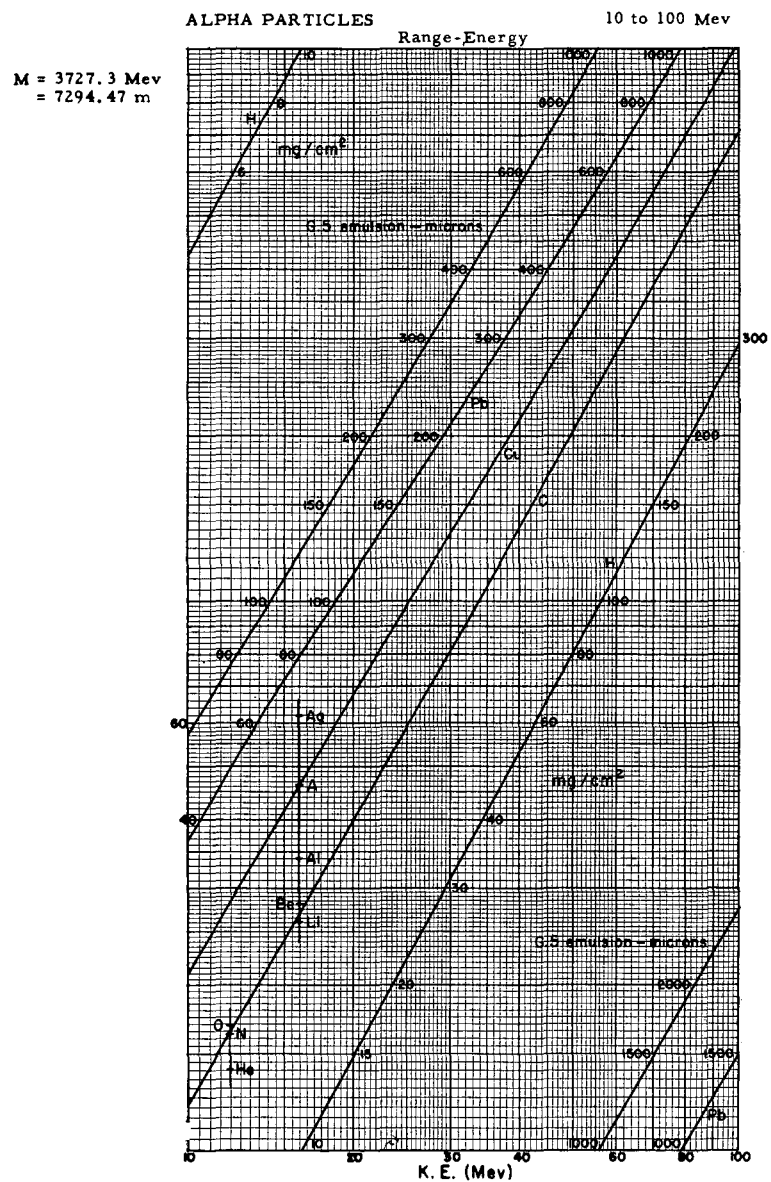


Fig. 38.

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