Title

SEPARATION AND ASSIGNMENT OF RADIOACTIVE ISOTOPES

Author

Michel, Maynard Cornelius.

Publication Date

1953-07-02

Thesis/dissertation

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

BERKELEY, CALIFORNIA

DISCLAIMER

UCRL-2267
Unclassified-Chemistry Distribution

UNIVERSITY OF CALIFORNIA

Radiation Laboratory

Contract No. W-7405-eng-48

SEPARATION AND ASSIGNMENT OF RADIOACTIVE ISOTOPES

Maynard Cornelius Michel

(Thesis)

July 2, 1953

Berkeley, California

TABLE OF CONTENTS

SEPARATION AND ASSIGNMENT OF RADIOACTIVE ISOTOPES

Maynard Cornelius Michel
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

July 2, 1953

ABSTRACT

A time-of-flight mass spectrometer of the type first developed by W. E. Glenn has been constructed for use as a separator for radioactive isotopes produced in cyclotron irradiations. This high transmission instrument allows sufficient mass separated activity to be collected to assign the mass numbers of such nuclides directly, in addition to providing pure samples of the given isotopes for the study of their decay properties.

Mass assignments and separations have been made on the following isotopes, some of which are reported here for the first time:

Isotope	Half-life
Cs^{125}	44 ± 1 min
Cs^{127}	6.2 ± 0.1 hrs
Cs^{130}	30 min
Tl^{198}	1.8 hrs
Tl^{198}	5.3 ± 0.2 hrs
Tl^{199}	7.4 ± 0.2 hrs
Tl^{200}	28 ± 1 hrs
Yb^{166}	58 ± 1 hrs
Yb^{169}	32 days
Tm^{165}	29 ± 0.5 hrs
Tm^{166}	7.7 hrs
Tm^{167}	9.6 days
Er^{160}	30 ± 0.5 hrs
Er^{161}	3.5 ± 0.2 hrs

SEPARATION AND ASSIGNMENT OF RADIOACTIVE ISOTOPES

Maynard Cornelius Michel
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California

July 2, 1953

I. INTRODUCTION

In research on the properties of an artificially produced radioactive nuclide, it is quite important to be certain of the assignment to a definite mass number and chemical element. Standard chemical procedures, if they can be applied within the time limit imposed by the half-life, are sufficient to establish the chemical identity, but less direct means are usually applied to the mass assignment. The most common of these are the method of cross bombardments and the study of the reaction yield as a function of the energy of the bombarding particle.

These methods have been quite consistent and are often adequate, but they sometimes are laborious or impossible to perform and interpret. And at best they offer an indirect approach.

The direct mass spectrographic assignment of mass numbers to radioactive nuclides has been rather common in the case of the naturally occurring activities and in the case of the uranium fission products, both cases in which weighable amounts of the isotope are available. In the case of the neutron deficient isotopes, usually formed by charged particle bombardments, one cannot normally produce such quantities.

Three methods have been used to make direct mass spectrographic assignments. Their success depends on the amount of material which

can be made (usually a function of the half-life) and on the chemical properties of the element.

The most general method is the detection of ions of the isotope in question by current measuring devices, just as in the case of the stable isotopes. This method is not often used except for those activities produced in large quantities by neutron capture reactions.

The second method, somewhat similar to that applied in the present work, is the use of high voltage electromagnetic separators, again only of use in the cases where large amounts of the isotope are available. The exception to this restriction is the case of elements which have gaseous compounds which are readily prepared. For these elements a gaseous arc source can be used which has a relatively high ionization efficiency for the many elements suitable for this source. Much work of this type has been done on the rare gases by workers such as Bergstrom.[1]

The third method is one used first by Hayden and Inghram for mass assignments of the uranium fission products.[2,3] Since this method is also used in this research in a preliminary survey of the rare earth isotopes, it will be described in some detail.

A conventional mass spectrograph, usually of the Nier type, is used with a low energy spread ion source which is capable of producing ions from solid compounds of the element in question. The ion spectrum is collected on a photographic plate where small amounts of the stable isotopes of the same element leave a line image of the source slit at the proper mass positions and therefore establish the mass scale. Usually the radioactive atoms which are collected will be in small

enough concentration to leave no direct image, but they will often make an exposure on standing merely from the radioactivity. More commonly, a second photographic plate, called a transfer plate, is placed face to face with the master plate and exposed for a period of time depending on the half-life of the activity. Development of this plate will now show images only where the radioactive atoms were collected on the original plate. It is quite obvious that successive exposures can give some indication of the half-life. This is often important if more than one active isotope exists in the same sample. Development of the original plate now establishes the mass scale, not known previously because of the difficulty of reproducing the magnetic field and other variables determining the mass. This method is of general usefulness and in the case where large amounts of activity are available, the decay can be followed directly on a Geiger counter, usually by mounting a counter behind a slit which can be passed over the photographic plate. Actual separation of the active isotope from the plate is not always practical since the mass scale is established by this master plate, which therefore must not be mutilated badly. This is not a very serious limitation, but a more direct physical separation is desirable. In addition, the transmission of a conventional magnetic mass spectrograph does not often exceed 2 percent of the ions formed, and it is often not possible to make sufficient activity to make mass assignments by this method, especially when rather large losses are taken in forming ions from the compounds of the element being investigated.

For this reason, any gain in ion transmission is of great importance in extending the sensitivity of this general method of making mass assignments, which is by far the most practical one yet devised for the limited quantities of the neutron-deficient isotopes usually made in cyclotron bombardments.

Of the other types of mass spectrometer known, only one, that first proposed by W. E. Glenn, was chosen as offering the best hope of reasonably good resolution at high masses combined with an increased transmission for ions.[4] The first machine of this type was built by Dr. Glenn in 1951 and was demonstrated to possess these desirable features. This research was undertaken to construct an instrument of this type, similar to the original design of Dr. Glenn, for use in separating usable amounts of relatively short-lived isotopes in order to assign their mass numbers and also to yield relatively pure samples for the study of their decay properties. The second section of this thesis describes the general features of this type of machine, and in particular the properties of the instrument constructed and used by the author, while the third section demonstrates the separation and assignment work which has been done on a selected group of isotopes in order to demonstrate its utility.

II. THE TIME-OF-FLIGHT SEPARATOR

A. Principle of Operation

The instrument is somewhat similar to the isotron[5] of Smythe and Wilson, but is designed for low accelerating voltages and has considerably better resolution.

A block diagram of the apparatus is shown in Fig. 1 where the upper part of the figure is a schematic representation of the analyzer tube.

Positive ions of the isotopes present in the sample are formed at A and accelerated through the dc field between the point A and the first bunching grid G1. Point A, all the plates B, and G1, are at an additional radiofrequency potential with a sawtooth wave form shown at the input to G1. This means that between G1 and G2 the ions receive an additional acceleration varying linearly with time and at a repetition rate of 2.2 megacycles. This sawtooth voltage can be adjusted to such an amplitude that ions of the desired mass will arrive at the point G3 in bunches, one period of the radiofrequency field apart in time, provided the distance G2-G3 is large enough to make the transit time long compared with the radiofrequency period. This can easily be seen since ions arriving late in a cycle receive additional acceleration, enabling them to overtake ions which have passed between G1 and G2 earlier in the cycle. The conditions for bunching are sufficiently non-critical that not only the desired mass but adjacent masses within several mass units will be well bunched, the degree of bunching decreasing rapidly beyond 10 mass units on either side, until those ions of mass far from the desired mass are not bunched at all. To meet the requirements for good bunching with reasonable radiofrequency voltages, the present machine, like the original one of W. E. Glenn, operates such that the transit time is 40 times the radiofrequency period.

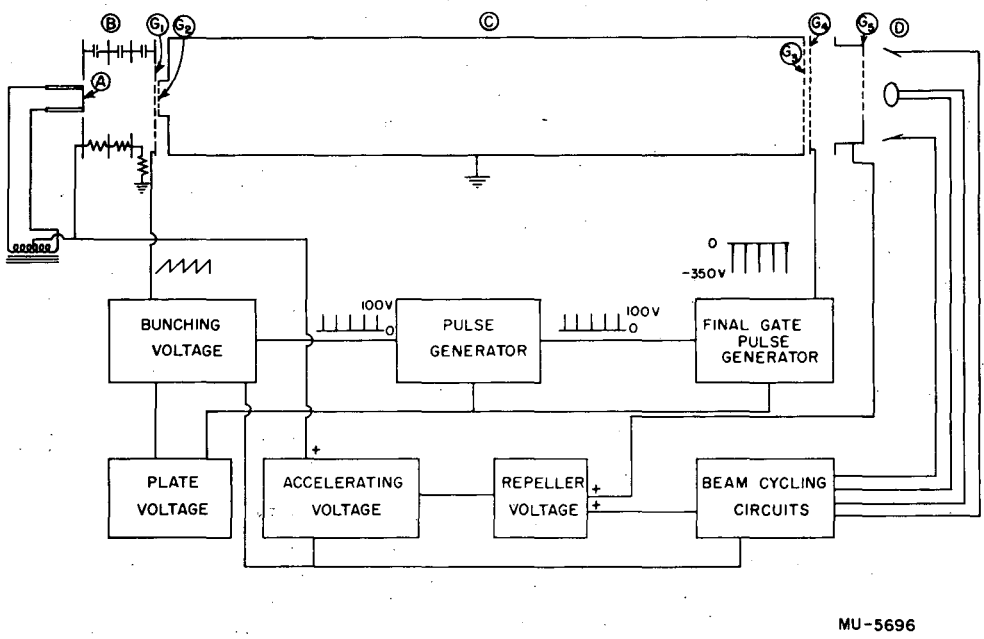

Fig. 1. Block diagram of time-of-flight separator.

Now that bunches of ions near the mass desired are arriving at G3, it will be noted that the time of arrival within a given cycle will be proportional to the square root of the mass of the ion. This achieves a separation in space depending on mass number. As indicated in Fig. 1, G5 is placed at a potential (variable) about 150 volts above the maximum value of the accelerating potential plus the bunching voltage. Thus no ions will be passed through G5 unless they acquire additional energy greater than 150 volts above their previous energy. This is accomplished in the gating grid circuit as ions pass between G3 and G4. In this region, coincident with the arrival of the desired bunch of ions, a sharp negative pulse of 350 volts amplitude is applied for about 0.01 microsecond. These ions which have been accelerated by this pulse can now pass through G5 and will strike any conductor near ground potential with roughly the full accelerating energy. Other ions, not receiving the extra pulse (which comes only once per radiofrequency cycle) are repelled down the tube by G5 and are eventually discharged on the tube walls.

Actually, rather than adjusting the time-of-flight, it is much easier to maintain the transit time and frequency constant and select the mass which is collected by varying the accelerating voltage and bunching amplitude. This does not change the principle of operation and considerably simplifies the calibration.

B. Choice of Tube Parameters

The fundamental choice which has been made in the two tubes built to date is the length of the analyzer tube (actually the distance

from G2 to G3) which has been chosen as about 27 inches. As mentioned before, the transit time has been chosen to be about 40 times the radiofrequency period in order to simplify the bunching requirements. The other parameters are fixed by these choices and the requirement that the accelerating voltages used be in a reasonable range, sufficiently high that the thermal energy spread of the ions be negligible for the desired masses, and not so high as to cause trouble from insulation problems and difficulty of bunching. The final choice of parameters is shown below for the second tube constructed.

Length of tube (L)	27.36 inches
Transit time (t)	18.2 microseconds
Repetition frequency (f)	2.20 megacycles
Accelerating voltage per mass unit (V/m)	7.48 volts/mass unit

The actual values of L and t and therefore of V/m need not be known accurately, since the instrument can easily be calibrated with a single known mass. Approximate values must be known only to be sure that one is operating such that the transit time is near the required value (collection is on the 40th cycle after the bunch leaves the source rather than the 39th cycle).

C. Operating properties

1. Mass Scale and Resolution.—With this system of operation, the accelerating voltage is directly proportional to mass (V/m is constant)

and therefore the overlap of adjacent masses is independent of mass (the resolving power ($M/\Delta M$) increases as the mass increases). This independence of resolution from mass is a decided advantage when working with relatively heavy masses as is often the case in the work for which this machine is intended. In fact, the amount of overlap does increase with decreasing mass number when one goes below about mass 70 because the initial energy spread of the ions from the source becomes an increasingly important factor in the resolution spread. For this reason, the present instrument has been built to operate only in the region of mass from 65 to 270.

The overlap of one mass on the adjacent mass is less than 1 percent of the peak intensity. This is shown in Fig. 2 which is a plot of the beam current due to Cs^{133} as a function of mass (or accelerating voltage).

2. Harmonics.—It can now be seen that at any given time, 40 different bunches of ions, each of the proper mass (and all other nearby masses), are traveling down the analyzer tube. The gating pulse which allows the ions in any particular bunch to be collected is the fortieth pulse arriving after these ions left the bunching grids. For this reason, it is important that adjacent undesired masses be well bunched or they would not be discriminated against completely, since it is possible for an ion of transit time 39/40 or 41/40 etc. of the proper transit time to be collected just as well as one of the proper time. And of course, a small number of any non-bunched ions (e.g., much lower mass impurities) will always get

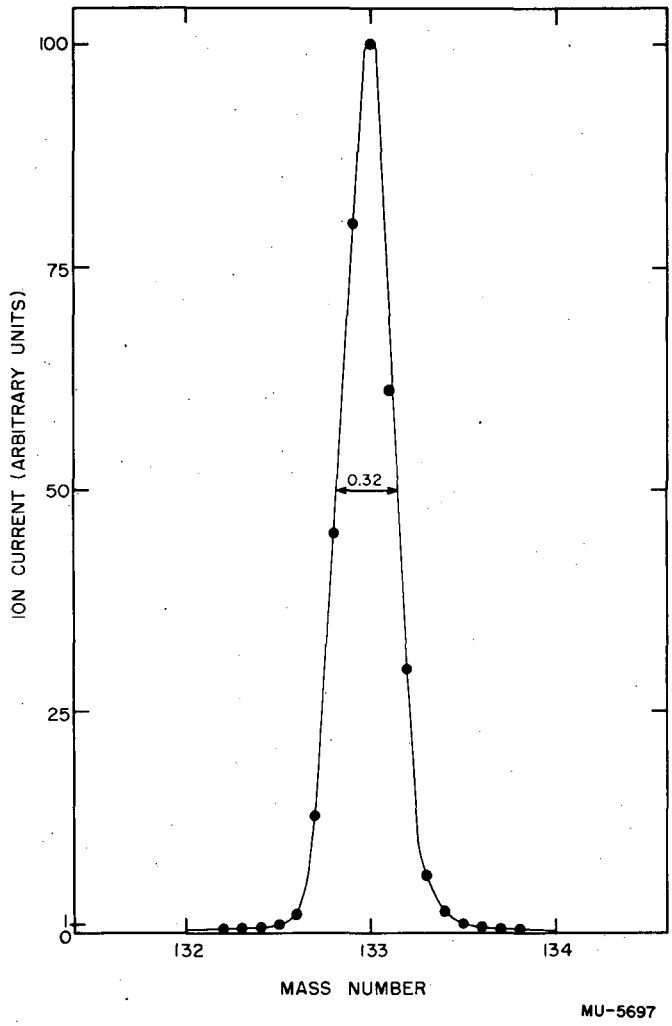

Fig. 2. Plot of ion current vs. mass number for mass 133.

through, being discriminated against only by the ratio of the width of the gate pulse to the length of the radiofrequency period. This presents a major difficulty in using this machine for isotope ratios, since impurities of such things as alkali metals of widely different mass from the desired sample will contribute a general background current which may fluctuate and obscure small peaks. Any practical method of eliminating these harmonics yet suggested reduces the transmission to approximately that of the conventional mass spectrometer.

It will be noted that for the collection of radioactive samples of isotopes of a single element, these harmonics will be no serious disadvantage, provided the range of masses present in the sample is smaller than the distance in mass units to the nearest harmonic from the highest or lowest mass present. This distance can be quite simply calculated and, for example, near mass 160, the effective mass spread of the nearest harmonics is about 8 mass units on either side (actually 7.9 mass units below and 8.1 mass units above 160). Thus if all isotopes in the sample are known to be within 6 or 7 mass units of each other, there will be no possibility of collecting one on a harmonic of its mass. In addition, the nearest harmonic will only accidentally fall on an integral mass number and would be severely discriminated against because of the rather sharp resolution. All the work in this thesis, as well as almost all similar mass assingment work will not be limited by this feature, since the spread of masses of the same element in a given bombardment is seldom over 6. In addition, one always has an idea of the relative amounts of each active isotope present and therefore the relative amounts which should be collected,

as well as an approximate idea of the mass range involved. Of course stable atoms of an impurity collected at the same time are of no consequence in the assignment of active isotopes.

3. Isotope Collection.—For the purpose of collection of active isotopes, the ions which have been selected according to a desired mass number are discharged on a platinum counting plate introduced in the collection end at ground potential. The majority of ions formed from the ion sources used are of the type M^+ and MO^+, which are non-volatile when discharged and remain as a thin uniform covering on the metal surface exposed to the beam. For calibration and checking the operation, stable ion beams can be observed by measuring the current produced by these ions when striking the collector, as in a conventional mass spectrometer.

In actual operation, only one mass can be collected at a given time, the others being repelled back down the tube. It is often a great time-saver to be able to collect several isotopes during one run and for this purpose the instrument has been equipped to switch automatically the accelerating voltage, the sawtooth bunching voltage and associated circuits, successively among 4 different mass numbers which can be pre-set to any desired values within 20 mass units of each other. A single switching cycle is complete within 15 seconds but the relative amounts of time for which each mass is collected are continuously variable from zero to 100 percent. Of course, the collection of more than one isotope from the same sample can only be done at the sacrifice of the total yield of each isotope, but it is

often justifiable to take this loss for the convenience gained. In the collector assembly there are arranged 4 separate collector plates which are simultaneously switched by the selector mechanism so that the same isotope is always collected on the same plate. During the part of the cycle that each plate is not being used to collect ions, it is placed at a positive potential sufficiently high to prevent collection of any ions which can be formed in the machine. For equal collection times the activity observed, corrected for the half-life and counting efficiency, can yield a rough isotope ratio. This serves as a check on the distribution of isotopes in a given sample which may be known from other considerations.

4. Transmission.--The primary advantage of this machine over conventional mass spectrometers is its unusually high transmission for ions. As stated before, a conventional machine rarely exceeds 2 percent, while the present time-of-flight separator is operating between 15 and 20 percent, as well as can be measured.

The transmission is here defined as the fraction of ions formed at the source which are eventually collected at the proper mass position. The primary sources of transmission loss are the lack of transparency of the grids, the lack of focusing in the source and incomplete bunching. The grids are each about 89 percent transparent and therefore account for a decrease of transmission to about 60 percent. In addition, the gating and bunching circuits have been determined to have a transmission of not greater than 75 percent. Experiments with source focusing have shown that a small increase in

transmission can be gained because at present not all the ions formed by the source pass through the bunching grids. However, the present linear field gradient is considerably simpler to use when the radio-frequency voltage must also be applied to the same electrodes and the gain to be achieved in transmission has not been considered worthwhile. It is believed that the use of finer grid wires, which has been attempted only recently, and other refinements such as source focusing can raise the total transmission to about 40 percent, but higher values will not be easily realized.

Measurements of the transmission referred to involve measurements of the ion currents to various parts of the machine while a steady beam of a single isotope is being accelerated. The reliability of these measurements is dependent on the number of secondary electrons emitted by the positive ions at the various surfaces. In addition, the measurement of total ion emission from the source is not easily made under conditions identical with the operating situation. For this reason the measured value depends on the assumptions made about the accuracy with which the total emission is measured. The actual transmission is given as a range of 15 to 20 percent, but no more precise value can be stated.

Upon the completion of the second analyzer tube it became desirable to check the total transmission of this tube for an active sample. The obvious choice is a long-lived cesium isotope, since it would require the least tracer material for the experiment. Because of the availability, Cs^{137} was chosen. This transmission check duplicated the actual operating procedure, except that there was no

necessity to work quickly and a better job was done. A few samples, each of about 20,000 c/m of Cs^{137} were run from the thermal ion source as sulfate samples without added carrier. The collected material was counted under the same conditions as the original sample, the collection efficiency for cesium being the fraction of the active atoms recovered at mass 137. The various runs were not in good agreement, since it has been found that the total efficiency depends upon several factors such as purity, rate of heating the sample, etc. In all, the various runs were in disagreement by about a factor of three, the better runs giving about 5 percent efficiency, or one atom in every 20 placed on the source being collected at the correct mass number. This is still felt to be a lower limit and is therefore in fairly good agreement with the measured transmission for ions, provided one assumes an ionization efficiency for cesium of about 25-50 percent, a value which is not in conflict with other observations. Since the estimated value of the total efficiency for cesium for a Nier type spectrograph is about 1/4 to 1/2 percent, this illustrates the rather high transmission of the present machine.

5. Calibration and Stability.--The 3 determining variables for the mass calibration in this instrument are the length of the tube, the accelerating voltage and the radiofrequency. The latter two can be held constant quite easily and the accelerating voltage is easily measured to a high degree of accuracy, so that the instrument should be self-calibrating. In practice the length of the tube need not be

measured but one can calibrate with any known mass, by use of an element of known isotopic constitution. An approximate knowledge of the tube parameters will then allow operation on the proper transit time. The bunching voltage will also be linearly related to the accelerating voltage and is in fact automatically controlled by the high voltage supply after being adjusted for the calibrating mass. It has been found that the mass scale is not quite linear but deviates by 0.05 mass units over a range of 75 or 100 mass units. This does not sound serious, but it can reduce the transmission considerably and is corrected for in actual practice by a calibration check of the machine before each run, with the stable isotopes of the element being used. In fact in those cases where a small amount of carrier has been added to the active sample, it is possible to calibrate at intervals during the actual collection. This is done as a precaution to insure correct operation, but it has not been found to be indispensable, except with the original tube for a reason to be discussed.

The stability of calibration is quite good in general but on occasion a slight drift will be seen over periods of several days. During a period of a few hours the change in calibration is never detectible. Shifts of as much as 0.05 mass unit have been observed over periods of a week, so that routine recalibration is carried out before each run. This stability is quite satisfactory and, of course, is necessary in a machine which must be run without internal calibration for many samples.

6. Tube Charging.--One of the most annoying problems in the construction of the original analyzer tube was that for total beam currents above about 10^{-11} amperes the reflected ions repelled by the repeller grid (G5) would collect on insulating spots on the wall of the tube and build up sufficient charge to deflect the beam and therefore lengthen the time-of-flight. The observed effect would be a shift in calibration such that the desired mass always came at a higher acceleration voltage. The magnitude of this effect was a function of the beam current in the tube, which depended not only on the size of the sample ion beam, but also on the ion beam due to impurities of sodium and other alkalis, which are almost always present and, because of the higher ionization efficiency, can easily account for a much higher beam than the sample. The effect on the calibration is variable and quite appreciable, amounting to as much as 1/2 mass unit if care is not taken to keep the maximum beam current below a reasonable limit. In view of the resolution curve in Fig. 2 this kind of a shift, if minimized by careful operation, will not invalidate the mass assignment, but will seriously limit the total fraction of the material collected.

Several methods of correcting this difficulty were tried, including the insertion of charge neutralizing filaments in the drift tube. These electron-emitting filaments did correct the effect for low beam currents and therefore raised the limiting beam current somewhat, but the problem was still present and the technical nuisance of replacing the filaments, or having them burn out during operation, was always present.

The obvious solution to the problem was to replace the tube with a larger diameter tube with polished walls having no oxide coating. This takes advantage of the reduction in the coulomb force exerted on the beam by any charge on the walls. The original brass tube was 2 1/2 inches in diameter. The final selector tube was of polished stainless steel 4 inches in diameter and, as expected, the charging effect completely disappeared. It is now possible to use any size beam up to about one microampere with no calibration shift. It is believed that even higher currents can be accommodated but present ion sources are limited in their emission to this region.

During the time the second tube was under construction, some of the work reported in this thesis was done with the first tube. At this time it was necessary to be very careful to control the maximum beam in the tube by monitoring constantly. In addition, the total yield of separated material was usually much less than with the second tube, due to the quick drop in transmission if the calibration shifted even slightly off the resolution peak. Nevertheless, considerable work was done with this tube and will be reported in section III. It is interesting to compare the work with Cs^{125} which was done in both tubes, the original tube yielding separated samples of 1000 to 1200 c/m from a sample of the size that gave 18,000 c/m in the new analyzer tube. However, the total transmission for ions when properly calibrated was the same for the two tubes within a factor of two. Most of the work done with the original tube has been verified by being run on the new separator also.

D. Comparison with Conventional Mass Spectrometers

The primary advantage of this machine over the conventional spectrometer is the increased transmission for ions. This has been discussed in detail previously. This transmission has been obtained with some sacrifice in resolution although the resolution is still quite sufficient for most purposes.

There are other advantages built into this particular instrument of a type which make work with very short-lived isotopes more convenient, primarily involving the ability to change samples quickly and to operate at relatively high pressures because of the short tube and the low accelerating voltages. For short-lived isotopes the sample can be inserted and the collection begun within 5-10 minutes.

The direct separation feature, so that one can follow the decay and determine other radiation properties more conveniently, is also quite useful although not a specific property of this machine. The advance knowledge of the mass scale is a valuable feature, since most other instruments rely on establishing the mass scale from the sample itself. The utility of this feature is evident, for example, in running artificial elements where no stable isotopes exist.

On the debit side, only one isotope can be collected at a time, resulting in a loss in transmission or inefficient use of all the isotopes present, depending on which is sacrificed. For most of the work to which this machine can be applied, this is not often a serious limitation, although the combination of this and a conventional machine for a preliminary survey (as was done in the later work reported in this thesis) makes for greater convenience when several isotopes of unknown mass number are produced in the same bombardment.

As has been demonstrated, the problem of harmonics is of little significance although it must be carried in mind during any work. It is a calculable and reproducible effect which can easily be recognized and avoided except in unusual cases.

There is one serious problem which has arisen in the separation of the isotopes of many elements. Because of the geometrical simplicity of the separator tube which makes the high ion transmission possible, the transmission for neutral particles is also high. Thus, if the collector plate is placed in the direct beam, neutral particles vaporized from the sample can collect on it also. In the case of inefficiently ionizing elements, the number of neutral particles emitted from the thermal ion source is much higher than the number of ions. For this reason the collection plates are placed away from the beam axis and arranged so that the ion beam is deflected 90 degrees to reach them, while neutral particles, not being deflected, are collected on the cold walls. Even so it has been noted that sufficient neutral particles are scattered to the collection plates to give a background of the unseparated activity on each mass separated plate. It was found impossible to reduce the amount of the vaporized activity in the present tube below a certain limit, depending on the element and chemical composition, but re-design of the collector could reduce this source of background to a negligible amount, without serious reduction of ion transmission. With the elements which ionize at high temperature, this background material can be quite high but never interferes with the assignment of the proper activity to the correct mass. A check of this vaporized activity is made in each run by leaving one

collection plate connected to a high positive voltage at all times so that only neutral particles can be collected. Careful checks indicate that the four plates receive the same amount of this background material to within 10 or 15 percent and therefore the single background plate is a good indication of the amount of vaporized activity being collected by all the sample plates.

In the case of separating an isotope in low abundance from one in high yield, this can be a serious problem and must be eliminated or minimized before much work of this type can be done.

E. Ion Sources

In the course of this work one is limited in the amount of the active isotopes that can be made. For this reason, the increased transmission of this machine has been very valuable. Any further increase in total efficiency must come, in the main, from an improvement in the ionization efficiency of the material, which will require improved ion sources. As can be seen from the low accelerating voltages, the ions to be used in this machine must not have a distribution of energies from the source alone of greater than 0.1 volt, if that high. This puts a severe limitation on the types of ion source that can be used. In most of the work reported in section III of this thesis the simple thermal ion source commonly used in isotope ratio mass spectrometers was used.[6] This source is often highly inefficient, but its simplicity is a definite advantage for relatively short half-life isotopes. In this source, a compound of the element to be studied (usually one which converts to the oxide on heating) is

evaporated on a thin tungsten ribbon and this ribbon is heated electrically in the source region to produce positive thermal ions with a low spread in energy. The efficiency of this source is highly sensitive to both element and chemical form, being most efficient for the oxides of metals in general. For a given element the ionization efficiency (the fraction of atoms placed on the source which are ionized) is a strong function of the experimental conditions of purity, etc., but the efficiencies for different elements range from values approaching unity for rubidium, to 10^{-8} for some transition metals like molybdenum. Fortunately many elements are in the region of efficiency greater than 10^{-5} which makes separation work with a few million disintegrations per minute quite practical.

A modified bombardment source in which the solid sample is vaporized into an electron stream in a relatively field free region has been used with some improvement in yield. It is capable of considerable improvement and will undoubtedly be of use in the case of elements which are extremely inefficient on the thermal source. Its complexity has resulted in no general use being made of this source in the present work on short-lived isotopes.

A third type of ion source which has been used is designed to avoid the inefficiency associated with a thermal ion source when used at high temperatures. A source somewhat like this was first used in 1951 by Mr. F. L. Reynolds of this laboratory, but the present source is a modification of a multiple filament source suggested and used by Inghram.[7] A drawing of this source is shown in Fig. 3 where the standard tungsten ribbon filaments are shown as A,B, and C. The filament B is the ionization filament while both A and C are sample mounting filaments. In operation all three filaments are at the same potential as the mounting plate except for the low voltage alternating current passed through them for heating.

The principle of operation is that ions are formed at the surface of filament B by contact of neutral particles evaporated from the

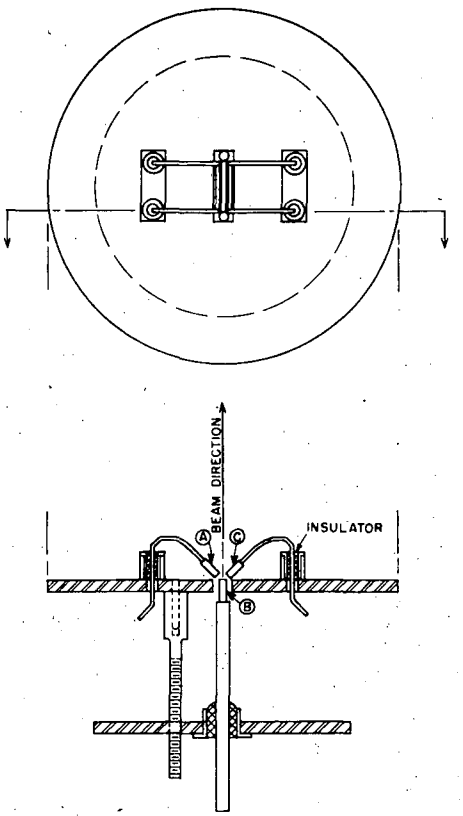

Fig. 3. Multiple filament ion source.

sample filaments. This type of source has been used successfully with the relatively poor thermal emitters, but it seems to be of much less value for relatively efficient thermal ionizers. Presumably, the advantage of this source lies in improving the efficiency of those elements whose ion-emitting compounds must be raised to such a high temperature for emission that vaporization as neutral particles is very much more probable than ionization, and quite rapid. The multiple filament arrangement allows a very high temperature (and therefore a higher probability of ionization) to be used in the ionization region while the rate of vaporization of the sample into this region is controlled by a much lower temperature. For elements where the thermal ionization efficiency is already high, not much is gained by this method, the small solid angle subtended by the hot filament becoming increasingly important. In fact, the ion yield may actually decrease below that for a conventional thermal source. This has been found to be the case for cesium. Even so, this source was of great value in the case of erbium, which is sufficiently poor as a thermal emitter that the multiple filament source adds to the yield significantly.

III. SEPARATIONS AND MASS ASSIGNMENTS

A. Cesium Isotopes

The first element chosen for isotope separation work was cesium, the choice being governed somewhat by the remarkably high ionization efficiency of that element from a thermal ion source. At the same time, Mr. Hirdaya Mathur of this laboratory was interested in the

lighter neutron deficient xenon and cesium isotopes and had already found evidence of a new cesium activity in this region. It was decided to look for this isotope directly on the time-of-flight separator in order to establish its mass number and correct half-life.

The cesium fraction was chemically separated from targets of CaI_2 which had been bombarded with 100 Mev helium ions. Since I^{127} is the only stable iodine isotope, the (a,xn) reactions are expected to produce all the neutron deficient cesium isotopes below mass 131. The gross decay of the cesium fraction showed periods of about 30 hours, 5.5 hours, and 45 minutes, the latter being the new activity. Cs^{129} and Cs^{127} were previously assigned mass spectrographically to the 31 hour and 5.5 hour periods[8] and the new activity was tentatively assigned to mass 125 on the basis of its half-life and its absence in bombardments at less than 50 Mev.

The chemical separation was made by dissolving the target material in water, carrying the cesium on silicotungstic acid precipitated from 13 M HCl, and redissolving this precipitate in water. This solution was passed through a short column packed with Dowex 50 cation exchange resin, which adsorbs only the cesium. The column was carefully washed with water and the cesium eluted with a little 13 M HCl. This was evaporated with a small amount of sulfuric acid and eventually placed on the tungsten filament of the standard thermal ion source. This separation gave a high yield of cesium activity relatively free of other alkali metal salts. Such impurities would have been undesirable even in small quantities, since the original separator tube was quite sensitive to large ion beams as explained in section II.

Cs^{125}.--Several collection runs were made on Cs^{125} (formed along with Cs^{127} and Cs^{129} in the bombardment of iodine with helium ions), a typical example being summarized below:

Mass number	Collection time (sec.)	Initial activity c/m
130	4	49
126	4	32
125	7	964
Bg.	-	15

This collection was made to collect three masses during the same run, by switching alternately from one to another of the mass numbers by means of the cycling device mentioned in section II. A fourth collection plate was kept at a potential such that only neutral atoms could be collected on it and the activity on this plate is labeled Bg. The column labeled collection time indicates the total time each mass number was collected during the 15 second cycle of the switching mechanism. The activity in column three is the initial counting rate of the sample plate, immediately after the run, in a chlorine-argon filled Geiger counter at about 10 percent geometry. This counting information applies to all the work reported in this thesis.

All three sample plates decayed rapidly and with approximately the same half-life indicating that most of the activity on the mass 130 and mass 126 plates was vaporized neutral material of about the same composition as the gross sample. The discrepancy between these

plates and the background plate was a little disturbing and several checks were made on the reproducibility of the amount of vaporized material on each plate. These experiments indicated that the plates were not in identically symmetrical positions with respect to the axis of the tube. This was corrected and steps were taken to reduce the relative amount of vaporized material in subsequent runs, especially in the second tube where the collection plates were placed farther from the tube axis.

Also, in this run the efficiency of collection of the Cs^{125} was not as high as had been expected, although this can be accounted for by a small calibration shift due to electrostatic charging of the tube walls. Several other collections were in agreement with this assignment.

After completing the second separator tube, a sample of Cs^{125} was collected from a similar bombardment in which the initial activity of Cs^{125} was 18,000 c/m with a background due to neutral atoms of less than 15 c/m. This sample was probably better than 99.9 percent pure and was used to study the gamma ray spectrum free from contamination by the other cesium isotopes, an experiment which is extremely difficult to do by any other method.

On the basis of the various separated samples, the half-life of Cs^{125} has been directly determined as 44 ± 1 minutes. This is in agreement with the 45 minute half-life obtained from a careful resolution of the gross decay. Cs^{125} was observed to decay with emission of a positron of maximum energy 2.05 Mev and an associated gamma ray of 0.115 Mev.[9] Details of the radiation characteristics will be reported elsewhere.

Cs^{130} and Cs^{129}.--Cesium^{130} has been reported as a product in low energy helium ion bombardments of iodine. It decays by emission of a positron with a 30 minute half-life.[8] This half-life offered some obstacle to direct assignment by means of a conventional mass spectrometer, but with the time-of-flight machine it was felt that the experiment was reasonable. Only the first separator tube was available at the time the experiment was undertaken and consequently the yields of separated material were quite low. Large samples of CaI_2 were bombarded with helium ions of approximately 23 Mev energy and the cesium fraction chemically separated and run in the time-of-flight separator. A typical example of these runs is shown in the following table.

Mass number	Collection time (sec.)	Initial activity c/m
131	5	2
130	5	130
129	5	14
Bg.	-	2

The mass 130 plate decayed with approximately a 32 minute half-life while the mass 129 plate had a decay which was in reasonable agreement with the reported 31 hour half-life for Cs^{129}, considering the small amount of activity collected. This isotope had previously been assigned with a Nier type mass spectrograph, and no further work was done on it.

Cs^{127}.--On several occasions, samples of relatively pure Cs^{127} were desired for study of the gamma rays emitted in its decay and for a direct determination of the half-life. The sample was chemically separated as described before from a target of CaI_2 bombarded with 75-100 Mev helium ions. This bombardment also produces Cs^{125}, Cs^{130}, and Cs^{129}. The first two decay out quickly, but considerable amounts of the latter are always present. One run was made in which three isotopes were collected. This run is summarized below.

Mass number	Collection time (sec.)	Initial activity c/m
130	5	9.5
129	5	28
127	5	498
Bg.	-	9

Since the Cs^{127} is (activity-wise) the most abundant isotope in the mixture, the separated sample of this isotope was quite pure. The decay curve was a straight line for over five half-lives until counting statistics began to scatter the data. On the basis of this and another larger separated sample, the half-life of Cs^{127} was found to be 6.2 ± 0.1 hours instead of the previous value of 5.5 hours. Again the Cs^{129} plate was followed and gave a half-life of 25-30 hours (after resolving out the background vaporized material) in essential agreement with the published value of 31 hours. The gross mixture was followed over 7 half-lives after the lighter isotopes had decayed and gave a half-life of 32.8 hours for Cs^{129}.

B. Thallium Isotopes

The bombardment of thick gold targets with 40 Mev helium ions gives rise to three activities of thallium which have been assigned to the mass numbers 198, 199, and 200.[10] The reported half-lives are 1.8, 7, and 27 hours respectively. All three decay by electron capture and the two longer lived isotopes have been studied rather thoroughly by other workers in an investigation of their radiation properties.[11]

A direct mass assignment on these isotopes was attempted early in the present work. The chemical separation was designed to separate the thallium fraction carrier-free from the relatively large amount of target material with a separation factor of better than 10^7, since it was found that small amounts of gold salts (1 microgram or less) had quite adverse effects on the ionization efficiency of thallium. The gold targets (10 grams) were dissolved in aqua regia and diluted slightly, the gold being reduced back to the metal with SO_2. It was found that this reduction could be made without appreciable carrying of the activity only if done under conditions where the gold was precipitated from a hot acid solution with excess chloride ion present. Under these conditions the gold is not always completely reduced. The remaining solution is oxidized with persulfate, adjusted to about 6 M in hydrochloric acid, and extracted with ethyl ether, which removes both the residual gold and the thallium in their (III) oxidation states. This ether solution is back extracted into 3 M HCl and passed through a 5 cm by 3 mm column packed with Dowex A-2 anion exchange resin, which adsorbs both the thallium and gold. After washing out most other contaminants with 0.5 M HCl followed by water,

the separation is carried out by reducing both the gold and thallium with a saturated solution of SO_2 in water, the thallium eluting off the column as Tl^+, and the metallic gold being held up in the body of the resin. The desired chemical state for the thermal ion source is the thallous sulfate, which is the product of this reduction. The sample, with a few micrograms of carrier is then evaporated on the ion source filament with a little excess sulfuric acid to insure conversion to the sulfate.

As previously noted, the gross decay can be resolved into three components, of about 1.8 hours, 7 hours, and 27 hours. Several collection runs were made in order to establish the mass numbers of these activities, but many of these experiments were of no value because of the sensitivity of the ionization of thallium to small amounts of impurities. The results of one of the more successful runs with the first separator tube are given below. This separation was finished about 10 hours after the bombardment, at which time the activity due to the 7 hour and 1.8 hour isotopes is in the ratio of 5 to 1.

Mass number	Collection time (sec.)	Initial activity c/m	Decay period
200	5	175	--
199	5	3883	7.3 hrs
198	5	1355	1.5 hrs 5.2 hrs
Bg.	-	200	--

The observed difference in activity on the background and mass 200 plates can be accounted for by the difference in counting efficiency of the two different Geiger tubes used for the measurements. This indicates the absence of any Tl^{200} which is not surprising, since it would not be expected to be present to the extent of more than 20 c/m on the basis of its abundance in the total sample.

The two periods observed on the mass 198 plate were a little surprising and it was felt this result might possibly have been the result of a partial breakdown of the automatic counting recorder used to follow the decay. For this reason it was felt necessary to prove the existence of the 5.2 hour isomer by a better experiment.

The proof of this assignment was deferred until the second separator tube was completed in order to take advantage of its greater stability. Then a similar bombardment was made in which the thallium fraction was allowed to decay for about a day after the bombardment, and the isotope separation was made about 30 hours after bombardment. In this case all the short-lived mass 198 activity should have decayed and only the longer-lived isomer would be present. The collection was arranged much as in the previous thallium separation as follows:

Mass number	Collection time (sec.)	Initial activity	Period observed
200	5	960 c/m	8 hrs 28 hrs
199	5	13,000 c/m	7.4 ± 0.1 hrs
198	5	2,500 c/m	5.3 ± 0.1 hrs
Bg.	-	89 c/m	—

This confirms the 5.3 hour activity at mass 198 which is believed to be an isomer of Tl^{198}, and confirms the assignments of the other two isotopes. The short period observed on the mass 200 plate is still not explained since one would expect at most only 100-150 c/m of the Tl^{199} to be present in this sample even if the resolution is as poor as a 1 percent overlap at the adjacent mass number. The observed amount of this short activity at mass 200 was nearly half the total sample (450 c/m). This is felt to be too much to be due to the resolution overlap and may perhaps be an indication of another isomer. Unfortunately the experiment necessary to prove or eliminate this possibility will be quite difficult to perform because of the difficulty of making large amounts of Tl^{200} free of Tl^{199}. This is not impossible and may be done in the future. At present, no serious claim can be made for the existence of the short-lived isomer of mass 200.

It will be noted that the half-life of Tl^{199} has been shifted to a longer value than obtained from the resolution of the gross decay. This is in agreement with the observation of a 5.3 hour period which cannot be resolved but yields an average value of 7.0 for both periods. In addition there is evidence that the short-lived isomer of Tl^{198} may have a half-life slightly shorter than 1.8 hours, although the certainty of this half-life, which is based on a much smaller sample, is less than for the other three periods.

Since in every case the chemical separation takes considerably longer than the half-life of the shorter isomer of Tl^{198}, there would be the possibility that the 5.3 hour period is really an isomer of

Hg^{198} not previously reported. However, from a consideration of the relative ionization efficiencies of thallium and mercury, it is felt that this possibility is extremely remote.

While these separated samples were available, the gamma ray spectrum of each was determined with a sodium iodide scintillation spectrometer. The gamma rays of Tl^{199} and Tl^{200} have been observed previously[11] and are summarized below with the energies observed with the mass separated samples.

Isotope	Gamma energy (Mev) (See Ref. 11)	Observed gamma energy (Mev)
Tl^{200}	0.365	0.365
	0.577	0.578
	0.622	0.610
	0.829	--
	1.210	--
	1.360	--
Tl^{199}	0.049	--
	0.078	--
	0.103	--
	0.157	0.150
	0.206	0.203
	0.245	0.240
	0.332	0.334
	0.454	0.454

The lower energy gamma rays not listed in column three for Tl^{199} were probably not observed because of a high conversion coefficient. The values in column two are from observations of the conversion electrons.

The separated Tl^{198} sample containing only the 5.3 hour isomer showed a clean spectrum with a prominent gamma ray at 0.405 Mev and a less abundant transition at about 0.650 Mev. No evidence of any gamma rays were seen which could be attributed to Tl^{199} on this plate.

The higher energy gamma rays of Tl^{200} were not searched for because of time limitations and the low activity of the sample. It is interesting to note that a careful search in the region below 500 kev shows no sign of the Tl^{199} gamma rays in this sample of mass 200. This is more indication that the short period observed on this plate may not be due to the overlap of the resolution in the separator. However any attempt to establish limits on the amount that could have been observed leads to the conclusion that this is not a conclusive argument in favor of the mass 200 isomer.

The assignment of the mass 198 isomer illustrates a case in which separation of isotopes by a method such as used here is able to make an identification which would be extremely difficult by most ordinary methods.

C. The Rare Earth Elements Ytterbium, Thulium, and Erbium

In the course of the work of Mr. W. E. Nervik on the spallation of tantalum by 340 Mev protons, it became necessary to know a little more certainly the mass numbers and decay properties of the neutron deficient isotopes of the heavy rare earth elements lutetium, ytterbium, thulium, and erbium, since these account for a considerable part of the spallation yield. It will be noted from Fig. 4 that few of the lighter isotopes of these elements were well accounted for. This

Lu							Lu^{170} B E.C. 1.7 d	Lu^{171} B E.C. 8.5 d	Lu^{171} D E.C. ~600d	Lu^{174} B E.C. 6.7 d	Lu^{172} B β^+E.C. 4.0h	Lu^{173} B E.C. ~500 d	Lu^{174} A E.C., β^- 165 d
Yb				Yb^{166} D E.C. 62 h		Yb^{168} stable 0.140 %	Yb^{169} A E.C. 31.8 d.	Yb^{170} A I.T. 1.6×10^{2}	Yb^{170} stable 3.03%	Yb^{171} stable 14.31 %	Yb^{172} stable 21.82 %	Yb^{173} stable 16.13 %	
Tm					Tm^{166} B E.C., β^+ 7.7 h	Tm^{167} B E.C. 9.6 d	Tm^{168} A E.C.β^- 85 d	Tm^{169m} A I.T. 6×10^{2}	Tm^{169} stable 100%	Tm^{170} A β^- 129 d	Tm^{171m} B I.T. 2.5×10^{2}	Tm^{171} B β^- 480 d	Tm^{172} E 2-3d.
Er	Er^{160} D β^+ ~17 h	Er^{162} stable 0.136 %	Er^{163} D β^+ ~65 h	Er^{164} stable 1.56 %	Er^{165} A E.C. 100 h	Er^{166m} A I.T. 1.7×10^{2}	Er^{166} stable 33.41%	Er^{167} stable 22.94 %	Er^{168} stable 27.07 %	Er^{169} B β^- 9.4 d	Er^{170} stable 14.88 %		Er^{171} B β^- 7.5 h
Ho	Ho^{160} C E.C., β^+ 22.5 m	Ho^{161} B E.C. 4.6 h	Ho^{162} B E.C. β^- 65.0 d	Ho^{163} B E.C. 5.20 d	Ho^{164} A β^- 34.0 m	Ho^{165} stable 100 %	Ho^{166} A β^- 27.3 h	Ho^{166} B β^- >30 y					

MU-5699

Fig.4. Summary of previously observed isotopes in the upper rare earth region.[12]

is due primarily to the difficulty of achieving a chemical separation of the rare earths from each other in a sufficiently short time to allow the study of any half-lives of the order of a few hours. The scarcity of bombardment target materials is also a factor in the lack of data on these elements.

Due to the efforts of Mr. Nervik, who collaborated in this work, a quite satisfactory chemical separation has been developed which allows separation of the four elements of interest 8 hours after the bombardment. This is sufficient to allow the observation of half-lives as short as 1 hour, although no mass separations were done on isotopes with half-lives less than 3 hours.

The basis of the chemical separation (which will be published in detail elsewhere) is an ion exchange method. The total rare earth fraction from the bombardment is adsorbed on a small amount of Dowex 4 percent cross-linked cation exchange resin and placed on the top of a 40 cm by 1.5 cm column packed with the same resin. Elution of the sample with pH 3.0 ammonium lactate solution produces the desired separation. The entire column is heated to 80° C by trichloroethylene vapor circulating about the column which is thermally insulated by a vacuum jacket.

A typical elution curve is shown in Fig. 5 in which the activity eluted per unit volume is plotted (upper curve) as a function of the time of elution, and the element eluted is labeled. The amount of each element eluted as determined by optical spectrographic analysis of stable isotopes added as carrier is plotted in the lower curve in order to verify the assignment of each activity peak to a given

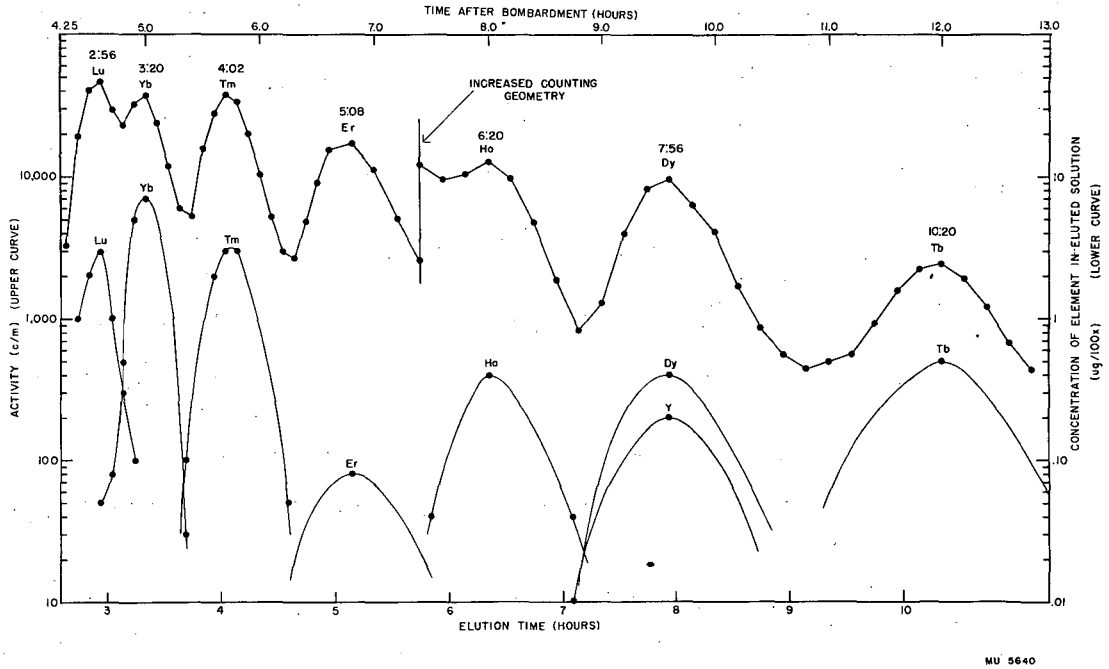

Fig. 5. Elution curve for rare earth separation.

element. The particular bombardment from which this curve was obtained was one of many in which metallic tantalum was bombarded with 340 Mev protons. The upper time scale is in hours after bombardment to give an idea when the various mass separations could be begun, while the lower time scale indicates the elution time after the beginning of the column run. Times to the peaks for each elements are labeled above those peaks.

Since the isotopes of these elements produced in the spallation of tantalum were of primary interest, this type of bombardment proved to be the most practical method of producing the desired isotopes, since it was demonstrated that sufficient activity could be produced by using relatively thick targets and bombardment times of 2-3 hours. Considerable quantities of activity are needed since these elements are not highly efficient thermal ion emitters, erbium being among the worst in the entire rare earth series, while lutetium has as yet not yielded usable results because of the difficulty of producing ions from any compound tried so far. In addition to the other obstacles, the temperatures at which these elements emit ions are sufficiently high that the vaporization of neutral particles from the ion source becomes an increasingly important problem which severely limits the sensitivity of detection of isotopes present in a sample in low yield. In general good results were obtained with sulfate samples containing several micrograms of stable element and 10^6 or 10^7 c/m of each isotope.

It was found by observing the gross decay of each separated element that several new decay periods had been formed, and some

previously reported isotopes were not immediately apparent. From the complexity of the decay curves this latter result was not surprising. One of the advantages of this method of producing the isotopes desired was that sufficient activity could be made without having large amounts of stable rare earth elements present (e.g., as target material) and consequently a faster chemical separation was possible.

The spallation yield for holmium is relatively low and no work is reported for this or lower atomic numbers.

Since the region under consideration contained several new isotopes as well as the possibility of mis-assignments of previously observed isotopes, it was felt that much time could be saved by making a preliminary survey of the entire mass spectrum of each element with a conventional Nier type magnetic deflection mass spectrograph. This was planned in order to observe at least the most abundant active isotopes which, now that the mass numbers were known, could be collected in the time-of-flight separator with a minimum of wasted effort. The observation of the decay of the separated samples furnishes the complete assignment. Of course relative half-lives can be obtained from the transfer plate technique and this data was of use in planning the mass separations.

The lower transmission of the conventional mass spectrograph for ions is partially amended by the relatively high sensitivity of the photographic transfer technique, especially for electron capture activities, which comprise the bulk of the isotopes under study. In addition, the mass separated samples from the time-of-flight separator were counted on a Geiger tube, with quite low counting efficiency for

these activities. These two facts tend to lessen the apparent difference in sensitivity of the two instruments. A more realistic use of the increased transmission of the newer instrument would have been made if the maximum counting sensitivity had been used. However, in view of the problem of the neutral background this was not felt justified since the vaporized material sets the real limit of sensitivity.

All the rare earth elements were used in the form of sulfate samples on the ion source. The standard thermal ion source was used on thulium and ytterbium, while a considerable improvement in yield was obtained in the case of erbium by the use of the modified multiple filament source described in section III.

1. Ytterbium.--A glance at Fig. 4 will show that ytterbium had only two previously known isotopes in the neutron deficient region, Yb^{166} (62 hours), and Yb^{169} (32 days). The gross decay of the ytterbium fraction from the spallation of tantalum shows a complex decay which can be resolved into components of the following periods:

Half-life	Isotope (probable)
78 min decay	Yb^{167}
7.7 hr growth	Tm^{166}
62 hr decay	Yb^{166}
32 day decay	Yb^{169}

The shorter period is a previously unreported isotope which has been assigned to Yb^{167} by chemical identification of its decay product (Tm^{167}) which has subsequently been assigned directly.

A sample of the gross ytterbium fraction was run in the Nier type mass spectrograph with photographic collection and using the photographic transfer technique described in section I. Transfer lines were obtained at mass 166 and mass 169 at intervals, the relative intensities varying in a way that is consistent with the assignment of the two activities to the 62 hour and 32 day periods respectively.

The same type of samples were run on the time-of-flight separator. The results of two runs are given below (all subsequent collections are made for equal times on each mass number).

Run I

Mass number	Initial activity c/m	Observed decay
166	450	7.4 hr growth 59 hr decay
165	100	--
167	100	--
Bg.	100	--

Run II

166	1977	7.5 hr growth 58 hr decay
169	681	31 day decay
Bg.	135	--

The half-lives reported in this and the following work were obtained from the total decay of the mass separated samples by simple resolution of the periods observed. Because of the background of vaporized material, each sample was not a straight line, but had more or less impurity of the other isotopes of the same element present in the original sample. In principle, a straight line decay should result (except for the growth of a daughter) by subtracting the background plate activity from that of any sample at various times. In practice, resolution of the curves is found to be simpler, but a careful check is made to see that the resolved activities, other than the sample being collected, agree with the amount of background actually present, due account being taken of the fact that some of the background material is also the same as that of the separated sample. In this way it is felt that whenever reasonable amounts were separated, the observed half-lives are a least as good as, and probably better than those obtained from the resolution of the gross decay curve, especially when the gross decay curve is complex. The one exception to this is the case of the longest-lived components of each element, such as Yb^{169}, where large pure samples for half-life determinations can be obtained by allowing the shorter components to decay out.

The failure to observe any significant activity at mass 165 and mass 167 merely confirms the belief that these two isotopes are either not formed in comparable yields or have quite short half-lives. As mentioned above, there is independent evidence for the assignment of the 78 minute ytterbium period to Yb^{167}.

2. Thulium isotopes.--The thulium fraction from the spallation of tantalum decays with a complex curve indicating periods of 7.7 hours, 9.6 days, 29 hours, and 85 days, the latter being in extremely small yield. As seen in Fig. 4, the 85 day, 9.6 day, and 7.7 hour activities have been assigned previously to Tm^{168}, Tm^{167}, and Tm^{166}, respectively.

Two runs of the thulium fraction were made in the Nier type instrument with photographic collection. The transfer plates made at varying times indicated only two active isotopes, at mass numbers 165 and 167. The relative half-lives were such that they could be assigned to the 29 hour and 9.6 day periods, respectively. No line was seen at mass 166 although these exposures were made within 12 hours after the bombardment, so that if Tm^{166} is not made directly, it would not have had time to form in maximum yield from the Yb^{166} parent, which has a 60 hour half-life. It has been noted from several observations, such as the low yield of shielded isotopes, that the primary method of formation of all the rare earth isotopes seen in these bombardments is by decay through the electron capture-positron decay chain, from an initial light tantalum or hafnium parent. This helps explain the low yield immediately after bombardment of such isotopes as Tm^{166} and Er^{165}.

Mass separation experiments were made on the thulium fraction at several different times after bombardment, the chemical separation being delayed until just before the mass separation. The result of two such runs are summarized below.

I. Chemical Separation 24 Hours After Bombardment

Mass number	Initial activity c/m	Observed period
167	460	9.6 days
166	718	8.0 hrs 53 hrs
165	690	29 hrs
Bg.	280	--

II. Chemical Separation Immediately After Bombardment

167	1258	9.2 days
166	490	8.0 hrs 50 hrs
165	1750	30.5 hrs
Bg.	190	--

The yield of the 7.7 hour thulium relative to the 30 hour activity was much higher in the case of the separation made a day after the bombardment, indicating that much of the yield is from the decay of Yb^{166}. The 50 hour activity at mass 166 is a little disturbing but it could easily be a small amount of Yb^{166}, which is made in very high yield, not completely separated by the chemistry. Not every chemical separation was entirely reproducible, so there is the possibility that perhaps as much as 2-3 percent ytterbium might have been in the cut taken for the thulium fraction. This much, coupled with

the fact that the ionization efficiency for ytterbium may be as much as five times that of thulium, can explain most of the 50 hour activity observed, which was less than one third of the total activity at mass 166. Of course, the yield of Tm^{166} in II is undoubtedly mostly due to the decay of Yb^{166} also, the time required for chemical separation being an appreciable fraction of the half-life of this activity.

The yields of any mass 163 or 161 thulium isotopes are quite low (less than one fifth of the yield of Tm^{165}) if present, and have not been observed as yet.

3. Erbium.--Erbium has presented the most difficulties, primarily because of the extremely low ionization efficiency with a thermal ion source. For this reason, the only significant results have been obtained with the multiple filament source described in section II. Even with this source, the total yield of erbium is less than with thulium, but of the same order of magnitude.

The gross decay of the erbium fraction from the spallation of tantalum shows decay periods of about 3.5 hours, 29 hours, and a long decay of about 9 days in very low yield. The two shorter periods are new isotopes and the 10 hour Er^{165} is not seen in this gross decay when chemical separation is made shortly after bombardment. However, a sample of chemically separated thulium can be milked for erbium to yield the 10 hour isotope, presumably as the decay product of Tm^{165}.

The erbium fraction was run on the Nier type mass spectrograph, again with photographic collection. The transfer plates showed three

active lines at mass 165, mass 161, and mass 160. The chemical separation here must be very good in order to avoid the possibility of a little thulium impurity overshadowing the erbium because of the large difference in ionization efficiency. It is believed that the line observed at mass 165 in this case is due primarily to thulium impurity, since no significant amount of activity can be collected as mass 165 in the time-of-flight separator under conditions where the chemistry has been more carefully performed. In this case the transfer plates indicated that the 3.5 hour activity and 29 hour activity could be assigned to mass 161 and 160, respectively. The line at mass 165 could be explained by a 29 hour half-life.

A summary of the result of a separation of the erbium isotopes made within 12 hours after the bombardment is given below. The samples were run as the sulfate on the multiple filament source.

Mass number	Initial activity c/m	Observed period
160	491	29.8 hrs
161	271	3.5 hrs
165	45	--
Bg.	24	--

The small amount of activity above background in the case of the mass 165 plate is not inconsistent with the amount of this isotope which might be present at the time of separation, but no real faith

can be placed in such a counting rate. The other isotopes collected were relatively pure and it is felt that the half-lives are reliable.

A point of some interest is the absence of an initial growth on the separated samples of Tm^{165}, Er^{160}, and Er^{161}. In the case of the erbium isotopes, the daughters have half-lives of 22 minutes and 4.6 hours, respectively, so that the sample used in the isotope separator had reached equilibrium before the mass separation. Thus, because of the higher efficiency of ionization of the daughter, the separated sample might well contain this daughter in roughly equilibrium amounts. If not, the mass separations in this case often took 2 or 3 hours, so that there was time for Ho^{160} to come to equilibrium during the run, while the Ho^{161} might well not be resolved from the 3.5 hour erbium even if it were present. The case of Tm^{165} showing no growth is more difficult to explain but the primary effect is probably that the counting efficiency of the Er^{165} is sufficiently low that no significant growth was seen on the sample which had come part way to equilibrium during the mass separation itself.

IV. CONCLUSION

The utility of the time-of-flight isotope separator has been demonstrated in the mass assignment of artificially produced radioactive isotopes and sufficient quantities of many isotopes can be collected to allow study of their radiation characteristics. This has been done with many of the samples collected in the course of the work done in this thesis, especially with the new isotopes which usually decay by electron capture. Here the investigation of the

gamma ray spectrum can be made easily with a scintillation counter and a pulse height discriminator.

The instrument has considerably higher transmission for ions than conventional mass spectrometers, a property of prime importance in cases where the amount of active material is quite limited. It has many other advantages which make it quite useful for relatively short-lived activities, where other methods of assignment or separation may also work, but become more difficult. Finally, since it is quite stable in calibration, it serves as its own mass standard and obviates the necessity for some independent establishment of the mass scale.

Further exploitation of the possibilities of the instrument may well be in the direction of increasing the ultimate sensitivity of detection by eliminating the present undesirable neutral background activity. A further increase in ion transmission can probably be made but any significant improvement in total yield will have to come from improved low energy spread ion sources for solid samples.

V. ACKNOWLEDGMENTS

Although it would be difficult to acknowledge the help of everyone who has contributed to this research, particular appreciation is expressed for the aid and guidance of Professor D. H. Templeton and for the helpful suggestions and criticisms derived from discussions with Mr. L. F. Tolman and Mr. F. L. Reynolds.

I am grateful for the assistance and advice of Dr. W. E. Glenn during the early stages of the construction of the instrument and for the contribution of Mr. Earl Hostetter in constructing the special grids necessary for the separator tubes.

The helpful cooperation of Mr. B. Rossi and his co-workers at the Crocker cyclotron, and Mr. J. T. Vale and the crew of the 184-inch synchro-cyclotron made the bombardments involved in this work possible.

This work was performed under the auspices of the U.S. Atomic Energy Commission. The author was the recipient of an Ethyl Corporation Fellowship during part of the research.

VI. REFERENCES

1. I. Bergstrom, S. Thulin, N. Svartholm, and K. Siegbahn, Arkiv f. Fys. 1:11, 281 (1949).
2. R. J. Hayden and M. G. Inghram, Phys. Rev. 70, 89 (1946).
3. R. J. Hayden, Phys. Rev. 74, 650 (1948).
4. W. E. Glenn, Ph.D. Thesis, University of California Radiation Laboratory Declassified Report UCRL-1628 (January, 1952).
5. R. R. Wilson, United States Atomic Energy Commission Declassified Document AECD-3373 (May, 1952).
6. J. P. Blewett and E. J. Jones, Phys. Rev. 50, 464 (1936).
7. M. G. Inghram and W. A. Chupka, private communication to F. L. Reynolds (March, 1952).
8. R. W. Fink, F. L. Reynolds, and D. H. Templeton, Phys. Rev. 77, 614 (1950).
9. H. Mathur, M. C. Michel, T. O. Passell, and E. K. Hyde, unpublished data (October, 1952).
10. D. A. Orth, L. Marquez, W. J. Heiman, and D. H. Templeton, Phys. Rev. 75, 1100 (1949).
11. H. I. Israel, and R. G. Wilkinson, Phys. Rev. 83, 1051 (1951).
12. J. M. Hollander, I. Perlman, and G. T. Seaborg, "Table of Isotopes," University of California Radiation Laboratory Unclassified Report UCRL-1928 (Revised) (to be published in Revs. Modern Phys., 1953).