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Momyer, Floyd F.

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STUDIES OF NEUTRON-DEFICIENT ISOTOPES OF EMANATION, FRANCIUM, AND RADIIU

F. F. Momyer

(Thesis)

February, 1953
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\[
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\end{align*}
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\[
\begin{align*}
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\]

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STUDIES OF NEUTRON DEFICIENT ISOTOPES OF
EMANATION, FRANCIUM, AND RADIUM
Floyd Franklin Momyer, Jr.
Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
February 1953

ABSTRACT

A search was made for new isotopes of emanation, francium and radium in the region of the 126-neutron closed shell.

The investigations of emanation activities were greatly facilitated by the development of a method for preparing emanation samples which could be counted by techniques typical for active non-gaseous samples. This technique consisted in brief of ionizing emanation atoms in a glow discharge and accelerating them into a wire or platinum plate through a potential of several hundred volts. Activities assigned to Em$^{211}$, Em$^{210}$, Em$^{209}$, Em$^{208}$, and Em$^{221}$ were discovered and their decay characteristics studied. The plot of alpha decay energy versus neutron number for emanation isotopes is strikingly similar to the plots for astatine and polonium isotopes at and below 126 neutrons.

Numerous experiments indicate that no low mass francium isotopes, other than Fr$^{212}$, exist with half-lives of greater than five minutes.

In bombardments of lead with carbon ions in the 60-inch cyclotron at Crocker Laboratory in Berkeley, a new activity assigned to Ra$^{213}$ was observed and its half-life and alpha particle energy determined. A sharp drop in alpha particle energy due to the 126-neutron closed shell is indicated in radium as in other elements down to bismuth.
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I. INTRODUCTION

Prior to the commencement of these studies, investigations of
neutron deficient isotopes of bismuth, polonium and astatine had
indicated a reversal in the trend of increasing alpha particle
energy with decreasing mass around neutron number 126 for these
elements.\(^1\) Alpha particle energies of isotopes of a given element
have been found to increase normally in a roughly linear fashion
with decreasing mass. However, in polonium, for example, there is a
sharp drop of about 4 Mev in alpha particle energy in going from the
isotope with 128 neutrons (Po\(^{212}\)) to that with 126 neutrons (Po\(^{210}\)).
This phenomenon is regarded as empirical evidence for the closed shell
at 126 neutrons, proposed theoretically by M. G. Mayer on the assumption
of strong spin–orbit coupling, with states of higher angular momentum
being favored.\(^2\) The alpha particle energies of Fr\(^{212}\) and of Em\(^{212}\)
produced from Fr\(^{212}\) by orbital electron capture had also been measured\(^3\)
and indicated that a sharp decrease in alpha particle energy also
occurred near 126 neutrons in francium and emanation. It might be
noted that emanation, rather than radon, is used herein to designate
element 86. Radon is considered the name of a specific isotope, Em\(^{222}\).
This is a question of nomenclature which apparently has not been
settled.
As a result of this afore-mentioned drop in alpha energy, a series of bismuth, polonium and astatine isotopes with neutron number 126 or less exist with half-lives long enough to permit chemical separation and study of their decay characteristics by means of the usual techniques. These nuclides have been studied extensively\textsuperscript{4-6} and the alpha particle energy versus mass number curve around 126 neutrons has been quite well determined for astatine and polonium. In bismuth, alpha half-lives are so lengthened at 126 neutrons that alpha branching in neutron deficient bismuth isotopes has not been observed for several neutron numbers below 126. Thus it was only by considerable effort and use of the powerful photographic emulsion technique that Dunlavey\textsuperscript{7} was able to observe and measure the energy of the alpha particle of Bi\textsuperscript{203}.

One purpose of this project was to look for any isotopes of radium of mass 214 or less with appreciable half-lives due to the stabilizing effect of the 126 neutron shell, thereby proving the existence of this region of increased alpha stability around 126 neutrons over a range of six atomic numbers (83 to 88). This would be convincing evidence that this phenomenon is the result of a closed shell at 126 neutrons. At the same time it was undertaken to search for new neutron deficient isotopes of emanation and francium in order to determine the shape of the alpha energy versus mass number curves at low mass numbers for these elements and compare with those for bismuth, astatine and polonium. Also, the nuclear properties of any new isotopes discovered were expected to be of general interest, quite apart from implications of nuclear shell structure.
As the atomic number increases from astatine, the region around 126 neutrons is becoming progressively more electron capture unstable, and the stabilizing effect of the closed shell is expected to increase alpha half-lives to only a few minutes at most for new isotopes of francium and radium. Thus it was expected that low mass isotopes of radium and of francium, other than Fr$^{232}$, which were long-lived enough to be separated chemically and studied would be few in number and that these would have half-lives of at most only a few minutes. It seemed likely that several new low mass emanation isotopes with appreciable half-lives might exist, however.

II. EXPERIMENTAL METHODS

A. Production of Activities

In most of this work, the activities to be studied were produced by bombardment of thorium foils with 340 Mev protons in the internal beam of the 184-inch cyclotron. It has been found that in this way neutron deficient isotopes of all the elements from lead to radium are produced in ample abundance by spallation. Th$^{232}$ was chosen as the target material as it is the first nuclide above the region of interest which is readily available and easily and safely handled.

It might be noted that prior to the construction of cyclotrons producing particles with energies of several hundred Mev, a project such as this would have been an extremely difficult, if not impossible, undertaking. The fact that no stable or very long-lived nuclides exist near the region of interest necessitates the production of these neutron deficient nuclides by spallation (i.e., requires high bombarding energies).
It is also noteworthy that a great variety of chemical species are produced in high energy proton bombardments by spallation and competing fission reactions. This in general complicates chemical procedures required for isolation of the element of interest. The procedures to be described later do not in general remove fission products with chemical properties similar to those of the element of interest. Since these fission products are beta emitters and the alpha decay properties were of main interest in this work, this was usually not a serious matter.

In some of the work another method of producing low mass emanation, francium and radium isotopes was found to have advantages. The 60-inch cyclotron at the Crocker Laboratory in Berkeley has produced sizeable internal beams of carbon ions (six times ionized) with energies in excess of 100 Mev. Earlier bombardments of gold foils in this beam had indicated that reactions such as $\text{Au}^{197}(\text{Cl}^{12}, \gamma 3n)\text{At}^{206}$ were occurring, with the principal yields being observed for the $4n$ and $6n$ reactions. Thus, bombardment of lead would be expected to produce low mass radium isotopes by such reactions at $\text{Pb}^{208}(\text{Cl}^{12}, \gamma 3n)\text{Ra}^{217}$. Emanation and francium isotopes of low mass number would result from the decay of these radium activities. Similarly, bismuth targets would yield low mass actinium activities which would be expected to decay very rapidly by alpha emission to francium isotopes of interest. The particular advantage of carbon ion bombardments is that it is possible by this means to produce isotopes of low mass number of an element free of any heavier activities. For example, in the case of radium, isotopes of mass greater than 220 were not produced in carbon bombardments.
As the use of carbon ions as bombarding particles is rather a new field, some details concerning the carbon ion bombardments performed in this work are discussed below.

It is possible to attain beams of $C^{+6}$ ions of the order of $10^{-7}$ amperes in the 60-inch cyclotron. The gross alpha activity in lead or bismuth targets at the end of a 10-minute bombardment is of the order of $10^6$ counts per minute. The majority of the particles in the $C^{+6}$ beam have energies in excess of 80 Mev. The energy distribution of the beam is continuous, apparently because the trajectories of the particles begin at various points within the tank instead of all at the center where the source is located. There also exists a beam, several times larger than the $C^{+6}$ beam, of $C^{+2}$ ions with energies of about 10 Mev. Targets were shielded from $C^{+2}$ ions by inserting a 19 mg/cm$^2$ tantalum foil in the beam. This stopped $C^{+2}$ particles but did not appreciably degrade the energy of the $C^{+6}$ ions.

It was necessary in bombarding lead and bismuth to take care that these metals were not melted by the beam. Even with the $C^{+2}$ beam eliminated the targets were occasionally destroyed by $C^{+6}$ ions. It was thus not possible to use the maximum beam intensity in bombardments (the magnet was generally tuned slightly off resonance).

In later bombardments of lead, it was found that lead could be quite firmly wedged to stainless steel plates by cleansing the steel surface with a special flux $^*$ and melting the lead onto the plate. The lead could later be dissolved off the stainless steel with hot $2 - 3 \text{ N HNO}_3$. A special probe designed by G. B. Rossi was used to

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*Ruby's Stainless Steel Soldering Flux, Ruby Chemical Co., Columbus, O.
hold such targets. The stainless steel plate could be mounted in this probe in such a way that a continuous stream of water cooled the back of the plate. The lead on the front of the plate was cooled effectively enough that it did not melt when bombarded with the maximum C\textsuperscript{6} beam. This probe had the disadvantage that there was no means by which the size of the beam could be measured directly. The cyclotron was thus tuned and the beam measured before each bombardment using another probe which was connected to an electrometer circuit. This probe was then replaced with the watercooled probe and the cyclotron run at the same settings as before. As the C\textsuperscript{6} beam appears to be somewhat unstable there was no assurance that the beam remained at the level previously measured throughout the bombardment. Such bombardments were usually satisfactory, however.

B. Techniques for the Study of Emanation Activities

1. Methods used in Preliminary Studies.—An instrument known as the differential alpha pulse analyzer has been used extensively in this laboratory in the study of alpha emitting nuclides.\textsuperscript{11} A thin (to avoid self-absorption) sample of the activity to be studied is placed on a plate and inserted in an ionization chamber. Pulses produced in this chamber due to ionization caused by alpha particles are applied to a linear amplifier. The output of this amplifier is proportional to the energy of the alpha particle over a range of several Mev. By applying the output pulse to a series of discriminator circuits, the pulse is counted on one of 48 registers according to the energy range within which it falls. The amplifier can be biased and the gain adjusted so that the 48 channels cover any energy range of
interest with a channel width of 15–20 kev or greater. With very thin
samples, the half-widths of mono-energetic peaks displayed on the
registers are about 50 kev. Absolute alpha energies are determined by
calibrating against two or more alpha particles of known energy in the
range covered by the 48 channels for given bias and gain settings.

At the outset, no simple method for obtaining alpha energies and
other decay characteristics of emanation isotopes of mass less than
212 was available. In the case of Em$^{212}$ it was possible to allow this
activity to grow into a sample of Fr$^{212}$ evaporated on a platinum plate.
In such a case most of the emanation will remain on the plate (being
entrapped in the solid material present) and the plate can be counted
on the differential alpha pulse analyzer to determine the alpha
class energy and half-life. Unfortunately, it has not been
possible to plate out a sample of parent activity large enough to
make this method feasible in the case of any emanation isotope of
lower mass.

In the early work, three methods of counting emanation activities
were used with some success.

The first of these had been used earlier by Meinke and Ghiorso$^{12}$
for counting the gross alpha activity in emanation samples from full
energy proton bombardments of thorium foils in the 184-inch cyclotron,
and also in studies$^3$ of Em$^{212}$ obtained from the decay of Fr$^{212}$. The
emanation sample to be counted is introduced, along with argon carrier
at one atmosphere pressure, into an evacuated brass ionization chamber
with charged central wire. This chamber is connected to a standard
amplifier and scaler used for alpha counting, and the alpha events
within the chamber counted. As some of the alpha particles produced
near the walls will produce most of their ionization in the walls instead of the gas, not all of the alpha events will be counted. The overall counting yield with the apparatus used was probably about 50 percent. Useful preliminary half-lives and mass assignments for alpha activities of which relatively pure samples could be prepared (Em$^{211}$ and Em$^{209}$) were obtained by using this technique to count a portion of such a sample and by studying the decay products from another portion of the same sample.

The second method was quite satisfactory for studying electromagnetic radiations from gaseous activities. The gas was condensed in a small glass bubble on the end of a piece of capillary tube by immersing the bubble in liquid nitrogen, and the capillary tube sealed off with a torch while the coolant was still on the bubble. The activity could then be counted on the gamma ray scintillation spectrometer$^{13}$ to determine the energies and half-lives of the radiations from the sample. The wall of the bubble could be blown thin enough that loss in intensity of electromagnetic radiations with energies of the order of 50 kev or higher was not serious.

The third method was useful in carbon ion bombardments. A thin layer of lead or bismuth (about 1 mg/cm$^2$) was volatilized onto a 1/4 mil tantalum foil. This foil was bombarded with the lead or bismuth facing away from the beam and with another 1/4 mil tantalum foil placed adjacent to the target material. Recoils from the target were thus collected on the second tantalum foil, which could then be counted on the differential alpha pulse analyzer or any of the other usual counting devices. Nuclei recoiling from C$^{12}$ collisions could have energies as high as several Mev so that emanation atoms
collected in the foil would be expected to be quite firmly affixed. This appeared to be the case. The first alpha particle spectrum analyses of emanation isotopes with mass number less than 212 were obtained in this fashion. The total initial alpha activity in such samples was of the order of a few hundred counts per minute and the resolution on the pulse analyzer was poor, due largely to beta-gamma activity induced in the tantalum foil itself. This resulted in straggling on the high energy side of the peaks, due to beta-alpha coincidences. Also, this method did not allow manipulation of the emanation so as to isolate and identify daughter activities and at the same time study the emanation activities present in order to make mass assignments, or in fact even to identify positively the element to which a given activity belonged. Nonetheless, valuable preliminary information on half-lives and approximate alpha energies was obtained in this way to guide future investigations. Tentative assignments of the activities observed were possible on the basis of alpha systematics.\(^1\)

2. The Glow Discharge Method.—After considerable preliminary work had been done using the above methods, a far superior technique was found. This new method made it possible to deposit emanation activities on a metallic foil or wire and to study the radioactivity so deposited in any manner applicable to normal non-gaseous samples. The technique is so simple and potentially so widely applicable to the study of other gaseous activities that it will be discussed here in some detail. It is similar in principle to that used by Bergström et al.\(^14\) for collecting samples of lower atomic number noble gases in the mass spectrograph.
The method was an outgrowth of some anomalous unpublished results obtained by W. E. Glenn, Jr. and E. K. Hyde during the course of runs in the time-of-flight mass spectrograph developed by Glenn.\textsuperscript{15} When Fr\textsuperscript{212} was run in this spectrograph, it was found that small amounts of Em\textsuperscript{212} from the orbital electron capture decay of Fr\textsuperscript{212} were collected on the receiver plates. Pursuing this observation, a glass tube was constructed with sealed in positive and negative electrodes, together with a heated filament. It was hoped that the filament would ionize some of the emanation atoms in a sample of emanation introduced into the tube and that the voltage across the electrodes would accelerate these ions into the cathode with enough energy that they would remain affixed there.

During the first trial runs the conditions of pressure and voltage within the tube were such that a glow discharge formed in the tube and emanation activity did collect on the cathode. In the presence of the glow discharge, it was found that the hot filament to produce ionization was quite unnecessary. The collection yield proved to be in the range of 5-10%, which was several orders of magnitude better than originally hoped for.

To determine half-lives and alpha energies satisfactorily, it was of course necessary that the emanation remain affixed to the plate after being deposited. It was found that no appreciable emanation diffused out of the surface of the plate at atmospheric pressure or in vacuum if the plate was not raised above room temperature. In a number of runs, the correct half-life of 3.8 days was determined for the decay of Em\textsuperscript{222} samples.

The samples appear to be quite thin as widening of the alpha
peaks due to self-absorption was not observed in the alpha pulse analyzer or in later runs on the magnetic alpha ray spectrograph, where half-widths of the peaks were about 10 kev.

Several slightly different tubes were later constructed and used. All consisted of a glass vessel with two sealed in electrodes arranged in various manners, and an outlet through which the emanation could be introduced. The emanation was collected on platinum plates attached to the negative electrode or on removable wires used as the cathode. The tube used in the later experiments and which appeared to give the best collection efficiency is shown in Figure 1. The electrodes were inserted through small holes in the end of the inner glass tube and sealed in with wax. A good seal which held well if not subjected to undue thermal or mechanical stress could be obtained in this way. The collector wire could be clipped off after each run, the seal loosened by heating, and more of the wire pushed through to serve as the electrode in the next run. Chromel or platinum wire was used, though there is no reason why the type of wire should be critical insofar as the collection process itself is concerned. The particular electrode arrangement shown was chosen as representing the best geometry for collection. Ionization should occur within the positive coil and all the positive ions produced are then subjected to a field converging on the negative wire.

The emanation to be collected is condensed in the tube with liquid nitrogen, the outlet closed, the liquid nitrogen removed, and the glow discharge run. The final pressure within the tube appears to result mainly from traces of oxygen.

As the main interest was in the application of this technique,
Fig. 1. Glow discharge tube used in the deposition of emanation activity on metal wires.
A little time was spent in studying the quantitative aspects of the collection itself. The glow struck over a range of pressures between 100 and 1,000 microns. A power supply providing dc voltages up to 1,000 volts was used. The voltage was adjusted to produce a stable glow with as low a current as possible (usually 2-3 milliamperes). The voltage required varied from 400 to 900 volts with variations in conditions within the tube from run to run.

A few early experiments on the collection efficiency were done using $\text{Em}^{222}$ obtained from known aliquots of $\text{Ra}^{226}$. As mentioned above, these indicated that the collection efficiency was a few percent in the case of the tube used (parallel electrodes with a platinum plate for collector). Actually, the efficiency appears to vary greatly with conditions in the glow discharge, so any numerical value has no quantitative meaning unless the geometry and pressure within the tube are fixed from run to run.

The discharge was usually run for a period of five minutes. Tracer runs seemed to indicate that the amount of emanation on the plate began to fall off after 10 to 15 minutes, probably due to gradual heating of the collector.

It was found that yields were consistently greater if collection was made in a series of short runs instead of one 5-minute run. This seems to indicate that the rate of collection is greater during the period immediately after the glow is struck. There are two possible apparent explanations for this. It is known that heating the collector will drive off the emanation affixed. The electrodes heat up during a run, however it is doubtful that an interval of a few seconds between successive glows would allow enough cooling to have much effect. A
more likely explanation is that an equilibrium condition is reached in the glow with a large region of high ion density and low potential gradient between the electrodes (the so-called plasma) and two small regions around the electrodes (sheaths) within which there are a relatively high gradient and low ion density. This situation is the result of a space charge of positive ions and a relatively smaller one of electrons around the cathode and anode respectively. The only region of very high gradient is confined to a small volume around the cathode. It is possible that the rate of collection is relatively very high at the instant of striking the glow and before this state is reached. If more nearly quantitative collection is ever required, it might be interesting to study the effect of producing the glow by applying a dc voltage with an ac ripple large enough to alternately strike and snuff the discharge superimposed.

At any rate, in the later experiments, a series of about ten 30-second glow discharges was run in place of one 5-minute one. Using the tube pictured and this technique, samples large enough for all our purposes and containing as many as $5 \times 10^8$ alpha disintegrations per minute of the mixture of emanation isotopes produced from thorium by spallation were obtained one hour after bombardment. This probably represents an overall yield of the order of 10 percent.

The vacuum system used for the emanation experiments is shown schematically in Figure 2. A Cenco-Hyvac pump and mercury diffusion pump were used in conjunction to obtain a base pressure in the system of between $10^{-6}$ and $10^{-5}$ mm of mercury. This allowed rapid and quantitative transfer of the gaseous samples from one trap to another.

In experiments involving direct removal of emanation from targets,
Fig. 2. Vacuum system used in the isolation of emanation fractions.
the target foil was placed in the flask A and a concentrated solution of sodium hydroxide above the sintered disk in B. Trap C was immersed in a dry ice-acetone mixture and traps D, E and F in liquid nitrogen. The system was evacuated and the dissolver solution (hot concentrated HCl with ammonium fluosilicate catalyst for the dissolution of thorium) allowed to drip slowly into A from the separatory funnel. The evolved gases were pumped off through B, C, D, E and F. A and B were then closed off and the system pumped down to base pressure. The emanation was next concentrated in trap F and transferred from there to the glow discharge tube. The discharge was then run and any emanation not collected was transferred back to trap F for use in possible later runs.

The sodium hydroxide bath was included to remove HCl from the evolved gases. The sintered disk insured the completeness of the cleansing operation by causing the gas stream to flow through the solution in the form of fine bubbles. Also, astatine has been found to behave as a gas at low pressure and room temperature and it was desired to remove this contaminant. In analogy to iodine, astatine disproportionates in solutions of greater than 0.1 M hydroxide ion. The hydroxide bath thus served also to convert any free gaseous astatine into non-volatile sodium compounds. The dry ice bath removed water vapor from the evolved gases and also served as further insurance against astatine reaching the system beyond. Actually the temperature of the dry ice-acetone mixture is about 10°C below the boiling point of emanation at atmospheric pressure, but at the pressures prevailing in the system, no appreciable emanation remained in this trap. The liquid nitrogen baths thus trapped emanation, along with krypton and
xenon present from fission and small amounts of oxygen present in the system. Three successive liquid nitrogen traps were used since due to the initial high pressure and high rate of flow of the evolved gases through the traps, not all of the emanation was condensed in the first.

In cases where it was desired to remove emanation from solutions of francium or radium, the solution of parent activity took the place of the hydroxide solution in B, and B was connected to a nitrogen line instead of to the flask A. The emanation was then swept from the solution by a slow stream of nitrogen gas, the rest of the procedure being the same as that outlined above. It might be mentioned before completing this discussion of the glow discharge method that H. B. Mathur of this laboratory has been quite successful in applying it to the study of xenon activities.\textsuperscript{17}

C. Chemical Procedures for the Isolation of Francium

The removal of francium from thorium was effected by using an improved procedure developed by E. K. Hyde.\textsuperscript{18} This involved coprecipitation of francium with free silicotungstic acid from cold, saturated HCl solution. To obtain carrier-free samples of francium, the silicotungstic acid precipitate was dissolved in water and passed through a Dowex 50 column. The francium adheres to the column and can later be removed with 6 N HCl. The procedure required about thirty minutes and resulted in carrier-free samples of francium which were radiochemically pure except for cesium fission product activities.

The daughters of low mass francium isotopes were investigated by studying the emanation activities growing into francium samples isolated within 5-10 minutes after bombardment by simple coprecipitation of francium with silicotungstic acid. The precipitate was dissolved
in water and pipetted into tube B (Figure 2.). The emanation was then removed by sweeping the solution with nitrogen as indicated above.

D. Chemical Procedures for the Isolation of Radium

In removing radium from thorium targets, the usual method of coprecipitation with BaCl₂ was used. The target could be dissolved in HCl (with ammonium fluosilicate catalyst added), the solution saturated with HCl gas and cooled in ice, and the BaCl₂ precipitate brought down within 5-10 minutes after the end of bombardment. The precipitation was made from about 10 ml of HCl using about 1 mg of barium carrier. Due to the presence of large amounts of higher mass radium isotopes and their daughters, it was not possible to pulse analyze such samples directly and obtain information on neutron deficient radium isotopes. However, the emanation daughters of such activities could be studied in the same manner as with francium, i.e., by dissolving the BaCl₂, transferring it to tube B (Figure 2), and later sweeping the emanation daughter activity from the solution.

A fast method was also developed for removing radium from lead targets bombarded with carbon ions in the 60-inch cyclotron. The lead was dissolved in a minimum amount of hot 2-3 N HNO₃ and the solution saturated with HCl gas and cooled in ice. In saturated HCl the lead forms a soluble chloride complex. Barium carrier (a few tenths of a milligram) was then added to precipitate BaCl₂ to carry the radium. With sufficient barium carrier and time to complete the precipitation, one would expect quantitative yields. However, the precipitation is not instantaneous and BaCl₂ is slightly soluble even in saturated HCl. To obtain thin samples for pulse analysis in a short time, incomplete precipitation of the barium chloride must thus be
tolerated and yields are sometimes as low as 20 percent, judging from tracer runs with Ra$^{226}$. Samples can be prepared in 5-10 minutes, but the time required for transport of the sample to the pulse analyzer, located at present a five minute drive from the 60-inch cyclotron, has made it impossible up to the present time to start pulse analyses sooner than about 15 minutes after the end of the bombardment.

III. EXPERIMENTAL RESULTS

A. Emanation

The greatest progress in these investigations has been made in the search for emanation isotopes of low mass number using the glow discharge technique. The isotopes of emanation with mass numbers 211, 210 and 209 were identified and their decay characteristics studied. In addition, an alpha activity tentatively assigned to Em$^{208}$ was observed. Samples containing all these activities plus Em$^{212}$ were run on the magnetic alpha ray spectrograph$^{19}$ in conjunction with F. Asaro to determine more accurate alpha particle energies. In this instrument, the energy separation of alpha peaks can be calculated to within one percent from their physical separation on a photographic plate placed at the end of their trajectory in the uniform magnetic field. Absolute energies can thus be calculated if the energy of one peak on the plate is known. Half-widths of the peaks for thin samples such as those used depend chiefly on the width of the source, since the magnetic field can be held quite constant during a run, and can be as low as 3-4 kev. The resolution of alpha peaks and accuracy of their measured energies are thus much better on this instrument than on the
alpha pulse analyzer. Alpha particle energies in this report are quoted to within 5 kev. The greater part of this uncertainty results from that in the energy of $\text{At}^{211}$, which was used as a standard. In these runs, the emanation was collected in the glow discharge on 10 mil chromel wire, which proved to be a suitable source for the spectrograph. Sufficient activity was obtained to permit changing the photographic plate several times during the run to obtain decay data on the alpha peaks which would confirm the assignments of the peaks.

In a slight digression from the main course of these experiments, some properties of the hitherto unobserved $\text{Em}^{221}$ were measured.

1. $\text{Em}^{212}$.—The half-life and alpha particle energy of $\text{Em}^{212}$ were determined before this work commenced as 23 minutes and 6.17 Mev respectively.\(^3\) During the course of this work, the energy of the alpha particle was redetermined on the pulse analyzer and found to be $6.23 \pm 0.02$ Mev. Finally, this energy was determined on the magnetic alpha ray spectrograph as $6.262 \pm 0.005$ Mev. Figure 3 is a plot of the results from a 15-minute exposure in the spectrograph started 65 minutes after the end of a 50-minute bombardment of thorium with 340 Mev protons. The half-widths of the peaks are about 10 kev. Successive exposures indicated that the peak assigned to $\text{Em}^{212}$ decayed with about a 23-minute half-life.

2. $\text{Em}^{211}$.—Early in these studies it was found that samples of emanation removed from thorium targets 24 hours after bombardment and counted on the gamma ray scintillation spectrometer exhibited a 16 hour decay in the combined astatine and polonium K X-ray peaks, after an initial growth. A growth and final 16 hour decay were also observed in the alpha activity of such samples when counted in the brass ionization
Fig. 3. Alpha spectrum of emanation fraction as determined on magnetic alpha ray spectrograph. Fifteen-minute exposure started 65 minutes after the end of a 50-minute bombardment of thorium by 340 Mev protons.
chamber. Traps which had held this activity for an appreciable length of time contained $^{211}\text{At}$ as identified by its chemical properties, measured 7.5 hour half-life and two alpha particles of energies, 5.85 and 7.43 Mev corresponding to $^{211}\text{At}$ and its $^{211}\text{Po}$ daughter. It thus appeared that $^{211}\text{Em}$ was an alpha emitting and orbital electron capturing activity of 16 hour period. This same activity appeared in samples of emanation swept from francium fractions obtained soon after bombardment, along with $^{212}\text{Em}$ and $^{210}\text{Em}$. As will be noted later, it is probable that this $^{211}\text{Em}$ is all or partly the result of direct contamination from the target and does not result from the decay of $^{211}\text{Fr}$ as was first thought.

These experiments were repeated later using the glow discharge technique. Samples of $^{211}\text{Em}$, pure of all but very small amounts of $^{222}\text{Em}$, were obtained directly from thorium targets a day after bombardment and studied on the 48-channel differential alpha pulse analyzer.

Pulse analysis immediately after preparation of the sample showed one peak of energy $5.82 \pm 0.02$ Mev. As time passed, another peak at 7.43 Mev ($^{211}\text{Po}$) was observed to grow in, indicating that $^{211}\text{At}$ was growing in from the electron capture branching of $^{211}\text{Em}$. Both of these peaks exhibited a final $16 \pm 1$ hour decay.

Figure 4 shows typical pulse analyses at 0.5 and 24 hours after preparation of the sample. The $^{211}\text{At}$ and $^{211}\text{Em}$ peaks were too close in energy to be resolved by the pulse analyzer, even with a channel width of 20 kev or less. However, after the growth of the $^{211}\text{Po}$ (7.43 Mev) peak was complete, the energy of the lower energy peak obtained by extrapolating the leading edge was $5.85 \pm 0.02$ Mev instead of the initial $5.82 \pm 0.02$ Mev. It appeared reasonably certain that
Fig. 4. Alpha pulse analyses of activity in initially pure sample of Em\textsuperscript{211}. Solid curve -- data taken 0.5 hours after preparation of sample. Broken curve -- data taken 24 hours after preparation of sample.
there was a real increase in the energy of the leading edge of the
peak, even though absolute alpha energies have a 20 kev uncertainty.
One would thus expect the leading edge of the double peak to have
the energy of At\(^{211}\), but the alpha particle energy of At\(^{211}\) had
previously been determined as \(5.89 \pm 0.02\) Mev.\(^6\) Samples of At\(^{211}\)
were therefore prepared and the energy redetermined on the pulse
analyzer. This redetermination did indeed yield \(5.85 \pm 0.02\) Mev as
the best value. A later determination of this energy on the alpha
ray spectrograph by R. W. Hoff and F. Asaro yielded a value of
\(5.862 \pm 0.005\) Mev.\(^{20}\)

From the ratio of the Po\(^{211}\) to the Em\(^{211}\)-At\(^{211}\) peak after
transient equilibrium was reached, from the half-lives of Em\(^{211}\) and
At\(^{211}\), and from the known branching ratio of At\(^{211}\), the branching
ratio of Em\(^{211}\) was calculated. The ratio of the high to the low
energy peak at equilibrium was \(1.0 \pm 0.1\). The branching ratio of
At\(^{211}\) had previously been determined as \(1.5\) (EC/\(\alpha\)) to within five per-
cent.\(^21\) The half-life of At\(^{211}\) was taken as 7.3 hours and that of
Em\(^{211}\) as 16 hours. A value of \(2.8 \pm 0.3\) for the orbital electron
capture to alpha particle emission ratio of Em\(^{211}\) results. The orbital
electron capture half-life of Em\(^{211}\) is then \(22 \pm 2\) hours and the alpha
half-life \(62 \pm 6\) hours. Using the above half-lives and branching
ratios, theoretical curves for the alpha activity in the Em\(^{211}\)-At\(^{211}\)
and Po\(^{211}\) peaks as functions of time elapsed after preparation of the
sample were constructed and compared with the empirical curves obtained
on the pulse analyzer. Figure 5 shows the fit of the empirical points
in one run to the theoretical curves.

The shape and half-width of the pure Em\(^{211}\) peak in alpha pulse
Fig. 5. Growth and decay of combined alpha activity in Em\textsuperscript{211} - At\textsuperscript{211} peak (unresolved), and growth and decay of Po\textsuperscript{211} alpha peak in initially pure sample of Em\textsuperscript{211}. Points are experimental. Curves are theoretical for the decay chain:

\[
\text{Em}^{211} \rightarrow \text{At}^{211} \rightarrow \text{Po}^{211}
\]
analyses at high gain settings (channel width 15–20 kev) suggested that the alpha spectrum of Em\textsuperscript{211} might contain two groups in comparable abundance with an energy separation of the order of 50 kev. It was beyond the capabilities of the pulse analyzer to determine whether this was definitely the case. However, in runs on the alpha ray spectrograph, two peaks identified as belonging to Em\textsuperscript{211} by their rough 16 hour periods were observed. Their energies were $5.847 \pm 0.005$ (35%) and $5.778 \pm 0.005$ (65%) Mev. A third peak of higher energy was observed to grow into the sample and so was assigned to At\textsuperscript{211}. The At\textsuperscript{211} peak ($5.862 \pm 0.005$ Mev) was actually used as an internal standard in these runs. Note that in Figure 3, the number of At\textsuperscript{211} tracks on the plate is less than half the number of tracks in the $5.847$ Mev Em\textsuperscript{211} peak.

This was a 15-minute exposure started about 20 minutes after preparation of the sample. In later exposures the At\textsuperscript{211} activity eventually grows to about five times that of the upper Em\textsuperscript{211} group.

The gamma ray spectrum of Em\textsuperscript{211} was studied on the gamma ray spectrometer. The growth and final 16 hour decay in the combined (unresolved) astatine and polonium K X-ray peaks (approximately 80 kev) has already been noted. Gamma rays of energies $65 \pm 10$ and $150 \pm 30$ kev in high abundance, and $400 \pm 30$ kev and $600 \pm 50$ kev in low abundance were also seen and assigned to Em\textsuperscript{211} by virtue of their rough 16 hour period.

In summary, the observed half-life of Em\textsuperscript{211} is $16 \pm 1$ hours, and the EC/a decay ratio is $2.8 \pm 0.3$. Two alpha groups of energies $5.847 \pm 0.005$ and $5.778 \pm 0.005$ Mev and respective abundances of 35 and 65 percent have been observed. Gamma rays of energies $65 \pm 10$, $150 \pm 30$, $400 \pm 30$, and $600 \pm 50$ kev have been assigned to Em\textsuperscript{211}. 
The $65 \pm 10$ kev gamma presumably results from the transition between the two states of Po$^{207}$ to which Em$^{211}$ decays by alpha emission. The genetic relationship of Em$^{211}$ to At$^{211}$ has been well established, so that this mass assignment is considered to be as good as that of At$^{211}$, which is to say quite certain. Figure 6 indicates the decay scheme for Em$^{211}$.

3. **Em$^{210}$**: An emanation activity with half-life $2.7 \pm 0.2$ hours and alpha particle energy $6.02 \pm 0.02$ Mev was also observed in pulse analyses of emanation fractions from 340 Mev proton bombardments of thorium. Figure 7 is a sample decay curve for this activity. This activity is probably to be identified with the 2.1 hour alpha activity reported previously by Meinke and Ghiorso in the gross alpha decay of such samples.$^{12}$

It was not possible to prepare pure samples of this activity, but in emanation fractions collected approximately four hours after bombardment, about 90 percent of the alpha activity belongs to this isotope, the remainder being Em$^{211}$ along with a few tenths of a percent of Em$^{222}$. After decay of the emanation, such samples were found to contain Po$^{206}$ and smaller amounts of Po$^{210}$ (no Po$^{208}$) as identified by their alpha energies of 5.22 and 5.30 Mev and half-lives of 9 days and roughly 140 days, respectively. The amount of Po$^{206}$ present indicated that the original samples must have contained a large amount of Em$^{210}$ alpha activity. The 2.7 hour, 6.02 Mev alpha activity was the only such unassigned activity present and was thus assigned to Em$^{210}$.

On the basis of this assignment, attempts were made to determine the percent alpha decay in Po$^{206}$ from the half-lives of Em$^{210}$ and Po$^{206}$.
Fig. 6

Decay scheme of Em 211.

Em 211
16 hr. EC

Po 207
5.7 hr.

5.847 (35%)
5.778 (65%)
Mev
a

At 211
7.3 hr.

EC

60%

5.862
Mev
40%

Bi 207
~50 yr.

EC

a

7.43
Mev

Po 211
0.5 sec.

Pb 207

Pb 203
52 hr.

EC

Tl 203

Em 211

16 hr.

EC
Fig. 7. Half-life of $^{210}$Em from decay of $6.02$ Mev peak on alpha pulse analyzer.
and the ratio of the initial alpha activity due to $^{210}\text{Em}$ to the amount of $^{206}\text{Po}$ alpha activity present after decay of the emanation. This is one of those too infrequent cases in which a branching ratio can be determined directly without involving rather uncertain estimates of yields or electron capture counting efficiencies. The $^{206}\text{Po}$ alpha activity amounts only to about 0.1 percent of the initial $^{210}\text{Em}$ activity and because of this fact such determinations are not so simple in fact as in principle. A peak containing more than a few hundred alpha counts per minute cannot be counted on the pulse analyzer with confidence in the accuracy of the observed counting rate. Thus one must either use samples with initial activity of this order of magnitude, or simultaneously follow the gross alpha decay of a large sample and pulse analyze a smaller portion of this sample to determine the fraction of the activity belonging to $^{210}\text{Em}$ at a given time. In this way the initial $^{210}\text{Em}$ activity in the large sample may be calculated and eventually the large sample itself may be simultaneously counted for alphas and pulse analyzed to determine the amount of $^{206}\text{Po}$ alpha activity present. In the first instance, the counting rate of the $^{206}\text{Po}$ after decay of the emanation is of the order of one alpha count per minute, so that the accuracy is limited by statistical error, background, and possible drift in the energy calibration of the pulse analyzer during long counts. In the second instance, more experimental quantities are involved in the calculations and this again lowers the accuracy. Four runs were made, two using the first and two using the second method. The extent to which $^{206}\text{Po}$ decays by alpha emission was calculated to be $5 \pm 1\%$. This average deviation is within experimental error and the actual value agrees with previous estimates of 5–10.
percent based on estimates of electron capture counting efficiency or of bombardment yields. The fact that consistent values for the alpha branching were obtained in this way serves to prove the genetic relationship of the 2.7 hour activity to Po\textsuperscript{206}. A serious error in the absolute value of the branching might result from loss of Po\textsuperscript{206} from the plate by alpha recoil. The samples collected in the glow discharge are quite thin and in vacuum one would expect 50 percent of the Po\textsuperscript{206} to leave the plate by recoil. However, in one case a platinum plate was suspended a few millimeters above the sample, held at a potential of (-) 2,000 volts with respect to the sample, and an attempt was made to collect recoils in air. The amount of Po\textsuperscript{206} on the recoil plate was only about 10% of that found on the sample. In the experiments, samples were allowed to decay in air in an enclosed space (in some cases within the pulse analyzer chamber). Possible error due to recoil loss has been neglected in the calculations and is probably substantially less than the limits of error placed on the percent alpha branching of Po\textsuperscript{206}.

The electron capture branching of Em\textsuperscript{210} can be calculated from the amount of Po\textsuperscript{210} present finally in the above samples. In only three of the runs was enough Po\textsuperscript{210} present to make reasonably accurate counting possible. In these the counting rate was a few counts per minute. The expected accuracy is low, especially since there is considerable difficulty in accurately resolving the Po\textsuperscript{210} from the initially twenty times larger Po\textsuperscript{206} peak. The average value of the EC/a decay for Em\textsuperscript{210} was 0.04 ± 0.01. It appears that Em\textsuperscript{210} decays by alpha emission to an extent greater than 95 percent. As recoils from electron capture have very low energy, there will be no
significant loss of the electron capture decay products of \( \text{Em}^{210} \) by recoil.

The question might arise as to whether the \( \text{Po}^{206} \) and \( \text{Po}^{210} \) observed in the samples could not have resulted from \( \text{At}^{206} \) and \( \text{At}^{210} \) present in the emanation fraction as contamination. Judging from the amount of \( \text{At}^{211} \) present initially in the samples, an upper limit of 0.1\% by activity can be set for \( \text{At}^{210} \) and \( \text{At}^{206} \) contamination, assuming equal cross sections for these three isotopes in the bombardment. This means that no appreciable amount of the \( \text{Po}^{206} \) observed could have resulted from \( \text{At}^{206} \) contamination and at most 10\% of the \( \text{Po}^{210} \) could have resulted from \( \text{At}^{210} \). The actual contamination is probably considerably less than 0.1\% as the amount of \( \text{At}^{211} \) observed represents just about the amount expected to have grown in from \( \text{Em}^{211} \) at the time of pulse analysis. It thus seems certain that the electron capture branching of \( \text{Em}^{210} \) has been observed and the genetic relationship of this activity to \( \text{Po}^{210} \) established.

Finally, the alpha particle energy of \( \text{Em}^{210} \) was determined on the magnetic alpha ray spectrograph as \( 6.036 \pm 0.005 \) Mev (see Figure 3). Successive exposures indicated that this peak decayed with approximately a 2.7 hour half-life.

In summary, \( \text{Em}^{210} \) has been identified by its genetic relationship to \( \text{Po}^{206} \) and \( \text{Po}^{210} \). The mass assignment is thus considered as good as the assignments of these two activities. The half-life of \( \text{Em}^{210} \) is \( 2.7 \pm 0.2 \) hours and the alpha particle energy \( 6.036 \pm 0.005 \) Mev. EC/a decay is less than 0.05 and the partial alpha and electron capture half-lives are thus greater than 50 hours respectively. The alpha branching of \( \text{Po}^{206} \) was determined to be \( 5 \pm 1\% \). Figure 8 shows the decay scheme for \( \text{Em}^{210} \).
Decay scheme of Em210

- Em210
  6.036 Mev
  2.7 hr. 
  a
  5.22 Mev
  5%  
  Po206
  9 d
  EC
  At210
  8.3 hr.
  EC
  P210
  138 d
  ~100%
  Po210
  5.3 Mev
  Bi206
  6.4 d
  EC
  Pb206

Fig. 8
Early in this work it was found that if emanation were swept from radium fractions removed from thorium targets within a few minutes after bombardment by 340 Mev protons, traps which had held the emanation fraction for 10-20 minutes contained radiochemically pure At\(^{209}\). At\(^{209}\) was identified by its alpha particle energy of 5.65 Mev and its half-life of 5.7 hours obtained both from alpha and beta-gamma activity decay. This was at the time considered evidence for an alpha emitting Ra\(^{213}\) with a half-life of a few minutes and an Em\(^{209}\) electron capture activity with a half-life of at least several minutes. With the advent of the glow discharge technique, one of the obvious experiments was to separate emanation daughter activity from radium fractions and subject this to the glow discharge in an attempt to obtain pure samples of Em\(^{209}\).

When this experiment was performed, it was found that pure samples of a new alpha activity were obtained. This activity was studied on the alpha pulse analyzer and found to have a half-life of 30 ± 2 minutes and an alpha particle of energy 6.02 ± 0.02 Mev (the same as that of Em\(^{210}\) to within experimental error). As the sample decayed, an alpha activity identified as At\(^{209}\) by its 5.65 Mev alpha particle and final 5.7 hour decay was observed to grow in. The fit of the empirical points to the theoretical curves for the amounts of Em\(^{209}\) and At\(^{209}\) activity as a function of time (5.7 hour daughter from a 30 minute parent) is shown in Figure 9.

The samples initially contained a few hundred alpha counts per minute of Em\(^{209}\) and the amount of At\(^{209}\) growing in was of the order of 10 counts per minute. If the branching ratio of At\(^{209}\) were known accurately, the branching ratio of Em\(^{209}\) could be calculated.
Fig. 9. Half-life of $\text{Em}^{209}$ from decay of 6.02 Mev peak on alpha pulse analyzer. Growth and decay of $\text{At}^{209}$ alpha activity (5.65 Mev peak on pulse analyzer) in initially pure sample of $\text{Em}^{209}$. Points are experimental. $\text{At}^{209}$ curve is theoretical for the decay chain:

$$\text{Em}^{209} \rightarrow \text{At}^{209} \rightarrow \text{Po}^{205}$$
Unfortunately, the At\textsuperscript{209} branching is not well known, but three runs were nonetheless made in an attempt to get some idea of the Em\textsuperscript{209} branching. For convenience, results were tabulated in the standard form of the ratio of the At\textsuperscript{209} alpha activity at five hours after preparation of the sample to the initial Em\textsuperscript{209} alpha activity. This ratio was found to be $1.3 \pm 2 \times 10^{-3}$. As any At\textsuperscript{209} resulting from the decay of Em\textsuperscript{209} during the preparation of the sample might be expected to collect along with the emanation if it were transferred into the glow chamber, there is some uncertainty as to where to choose the zero of time (the time at which one can consider At\textsuperscript{209} to start growing into an initially pure sample of Em\textsuperscript{209}). It was undertaken to minimize this error by transferring the emanation into the glow chamber from a dry ice trap which should retain any astatine present. The limits on the value quoted allow for the uncertainty in the zero time. No other appreciable errors are apparent if one makes the plausible assumption that astatine collects at most only as efficiently as emanation in the glow discharge. Calculations then yield an electron capture to alpha decay ratio of $5 \pm 1$ for Em\textsuperscript{209}, assuming 5\% alpha branching\textsuperscript{6} for At\textsuperscript{209}. The greatest uncertainty in this calculation is in the figure for the alpha branching of At\textsuperscript{209}. This would mean a partial electron capture half-life of $36 \pm 3$ minutes and a partial alpha half-life of $3 \pm 0.5$ hours for Em\textsuperscript{209}. This is about the value predicted as one expects the alpha half-life of Em\textsuperscript{209} to be greater than or equal to that of Em\textsuperscript{210} ($2.7 \pm 0.2$ hours). This consideration would set a lower limit of about five on the expected branching ratio. A rough cross check was also carried out later by observing the amount of Em\textsuperscript{209} growing into a sample of Ra\textsuperscript{213}. The branching ratio calculated in this case was $6 \pm 2$. The
greatest significance of the preceding is that the branching ratios obtained by assuming the Ra\textsuperscript{213}→Em\textsuperscript{209}→A\textsubscript{t}\textsuperscript{209} genetic relationship for the activities in question are consistent within themselves and compatible with other data and estimates. This increases confidence in the present assignment of the activities.

The smaller alpha branching of Em\textsuperscript{209} and the fact that its alpha energy is about the same as that of Em\textsuperscript{210} account for the fact that Em\textsuperscript{209} was not originally identified in pulse analyses of emanation fractions produced by spallation. However, once the properties of Em\textsuperscript{209} were known, the initial decay of the 6.02 Mev peak in samples counted starting about 30 minutes after bombardment indicated that Em\textsuperscript{209} alpha activity was 0.8 ± 0.2 times that of Em\textsuperscript{210} at the end of a 20-minute bombardment. This is approximately what would be expected if the alpha half-lives and cross sections for the two are about equal.

The Em\textsuperscript{209} alpha particle was not resolved from that of Em\textsuperscript{210} in the runs on the alpha ray spectrograph either, but assuming Em\textsuperscript{209}/Em\textsuperscript{210} alpha activity is 0.8 at the end of a 20-minute bombardment, one calculates that 1/7 of the tracks in the 6.036 Mev peak in Figure 3 belong to Em\textsuperscript{209}. The decay of this peak also seems to indicate the presence of an activity shorter lived than 2.7 hours, but the accuracy of the decay points is not good enough to allow accurate determination of the amount. Assuming that 15% of the alpha tracks in the peak are due to Em\textsuperscript{209} in this exposure, it appears that the alpha particle energy of Em\textsuperscript{209} is exactly equal to that of Em\textsuperscript{210} or is less by not more than 10 kev.

In summary, Em\textsuperscript{209} has a half-life of 30 ± 2 minutes and an electron capture to alpha decay ratio of about 5, assuming 5% alpha branching in
The alpha particle energy is $6.02 \pm 0.02$ Mev from pulse analyzer measurements, and from runs on the magnetic alpha ray spectrometer, appears to be $6.036 \pm 0.005$ Mev. The genetic relationship of this activity to $\text{At}^{209}$ has been well-established and the mass assignment is thus considered as good as that of $\text{At}^{209}$. The decay scheme for $\text{Em}^{209}$ is shown in Figure 10.

It might be noted here that when it was made certain that the radium fraction was clean, there was no evidence for any other emanation activities in the samples. In one case $\text{Em}^{212}$, $\text{Em}^{210}$, and $\text{Em}^{211}$ appeared. These were apparently contamination from the target solution judging from their relative amounts. It thus appears that $\text{Ra}^{214}$ and $\text{Ra}^{212}$ both have half-lives of less than a minute, as expected.

5. $\text{Em}^{208}$—In emanation fractions obtained from thorium by spallation, and affixed to metal plates by the glow discharge method, an alpha activity with energy $6.14 \pm 0.02$ Mev and half-life $23 \pm 2$ minutes was observed on the pulse analyzer. Figure 11 is a sample decay curve for this activity obtained from pulse analysis.

The fact that the half-life of this activity is the same as that of $\text{Em}^{212}$ to within experimental error would at first suggest that this might be an alpha group belonging to $\text{Em}^{212}$. Pure samples of $\text{Em}^{212}$ isolated from $\text{Fr}^{212}$ contain no such group, however. In spallation yields, the ratio of $\text{Em}^{212}$ activity to this activity at the end of bombardment was $4.7 \pm 0.5$ as determined from runs on the alpha ray spectrograph. (Correction was made here for the amount of $\text{Em}^{212}$ growing in from $\text{Fr}^{212}$ after the end of bombardment and before removal of the emanation from the target). In one instance a target of enriched lead (Pb$^{204}$ - 15%, Pb$^{206}$ - 50%, Pb$^{207}$ - 15%, and Pb$^{208}$ - 20%) was bombarded with carbon ions in the 60-inch cyclotron and recoils
Decay scheme of Em 209

- Em 209
  - 30 min. α
  - 1.5 hr. Pb 205
    - 6.036 Mev
    - 85%
    - EC
  - 5.65 Mev
    - 5.2 α
    - 99.4%
    - Pb 201
    - 8 hr.
    - Pb 204
  - 4.86 Mev
    - Bi 205
    - 14.5 d
    - EC
    - <10%
  - Po 209
    - 100 yr.
    - EC
    - α
    - Bi 209

Fig. 10
Fig. 11. Half-life of Em$_{208}^{208}$ from decay of 6.14 Mev peak in alpha pulse analyzer.
from the target collected on 1/4 mil tantalum for pulse analysis. In this case, the 6.26 and 6.14 Mev peaks were present in approximately equal abundance. It thus appears established that the 6.14 Mev alpha particle belongs to a new and distinct alpha activity.

Traps which had contained this activity for 10-20 minutes were found to contain Po$^{204}$ as identified by its alpha particle energy of 5.35 Mev and measured half-life of about four hours. The presence of an alpha emitting Em$^{208}$ in the sample was thus indicated. The 6.14 Mev activity was the only activity observed which was not otherwise definitely assigned and so was identified with Em$^{208}$. This assignment fits systematics well.

From the initial alpha activities of Em$^{208}$ and Em$^{212}$ in a sample and the final amount of Po$^{208}$, the electron capture branching of Em$^{208}$ can be calculated. Such a calculation yielded a value for the EC/a decay ratio of Em$^{208}$ of 4. This value is probably good to 25%, but the chief significance attached to it is qualitative and not quantitative. It appears certain that Po$^{208}$ from the electron capture decay of Em$^{208}$ is present in the sample. The assumption that the yield of Em$^{208}$ is equal to or less than that of Em$^{212}$ at the end of bombardment leads to an upper limit of 4 for EC/a decay (Figure 16). It is seen that the branching ratios obtained from these two different considerations are about the same. This lends credence to the assignment of the activity. The alpha half-life of 1-2 hours calculated for a branching ratio of the order of 4 is also reasonable for Em$^{208}$.

The alpha particle energy of Em$^{208}$ was determined on the alpha ray spectrograph to be 6.138 ± 0.005 Mev (see Figure 3). Three successive exposures indicated a half-life of 23 ± 3 minutes for the
decay of this peak.

In summary, $\text{Em}_{208}^{208}$ appears to be a $23 \pm 2$ minute activity with alpha particle energy $6.138 \pm 0.005$ Mev. There is some evidence for a genetic relationship between this activity and $\text{Po}_{204}^{204}$ and $\text{Po}_{208}^{208}$. The mass assignment is still considered somewhat tentative however, especially since the assignment of $\text{Po}_{204}^{204}$ is not a certainty.\textsuperscript{5} The EC/$\alpha$ decay ratio appears to be approximately 4. Figure 12 shows the decay scheme of $\text{Em}_{208}^{208}$.

6. $\text{Em}_{221}^{221}$—Although it was somewhat afield from the purpose of these experiments, it was decided to look for the hitherto unobserved $\text{Em}_{221}^{221}$ while all the necessary apparatus was at hand.

For this purpose, thorium was bombarded with 110 Mev protons and the emanation fraction removed from the target and collected in the glow discharge apparatus as usual. A typical pulse analysis of such a sample is shown in Figure 13.

The 6.08 and 6.33 Mev peaks correspond to $\text{Fr}_{221}^{221}$, the 7.02 Mev peak to $\text{At}_{217}^{217}$, the 6.14 Mev peak to $\text{Em}_{208}^{208}$, and the 6.26 Mev to $\text{Em}_{212}^{212}$. Pulse analyses at other settings definitely established the existence and counting rates of the 6.14 and 6.08 Mev peaks. Later an alpha peak of energy 8.34 Mev ($\text{Po}_{213}^{213}$) grew in, the growth and decay corresponding roughly to that expected for 47 minute $\text{Bi}_{213}^{213}$ resulting from the decay of a 25 minute parent. Peaks corresponding to $\text{Em}_{222}^{222}$, $\text{Em}_{210}^{210}$ and $\text{Em}_{211}^{211}$ were also observed. The 6.54 Mev peak appeared to be a new activity.

$\text{Fr}_{221}^{221}$ is a 4.8 minute and $\text{At}_{217}^{217}$ a 0.02 second activity. However, the 7.02 and 6.54 Mev peaks decayed with a $25 \pm 2$ minute half-life. Accurate counts on the $\text{Fr}_{221}^{221}$ peaks were difficult because of the $\text{Em}_{212}^{212}$
Decay scheme of Em 208

Em 208
23 min.
EC

Po 208
3.8 hr.
α
6.138 MeV
~80%

At 208
1.7 hr.
EC

Pb 204
18 hr.
EC

Pb 204

Bi 204
12 hr.
EC

Po 208
3 yr.
α
5.108 MeV
0.5%

Em 208
208

Fig. 12

00.
Fig. 13. Alpha pulse analysis of $^{221}$Em-$^{212}$Em sample.
present, but these appeared to decay with roughly this same half-life. Figure 14 shows sample decay curves for the 6.54 and 7.02 Mev peaks with some points for Fr$^{221}$ included. The statistical accuracy is not good, especially for the 6.54 Mev peak, but all these activities appear to have the same half-life of 25 minutes. One is thus led to the conclusion that the Fr$^{221}$ chain results from the decay of a 25 minute beta-emitting Em$^{221}$. The 6.54 ± 0.02 Mev peak is tentatively assigned to Po$^{217}$ as it decays with the half-life of Em$^{221}$ and has roughly the energy estimated for Po$^{217}$ from alpha systematics. At equilibrium, the ratio At$^{217}$/Po$^{217}$ is $5 \pm 1$. Correcting for the fact that the half-life of Fr$^{221}$ is appreciable with respect to that of Em$^{221}$ and assuming a short half-life and negligible beta branching for Po$^{217}$, a beta to alpha ratio of 4 ± 1 for Em$^{221}$ results. Any excess of At$^{217}$ over Fr$^{221}$ activity should in theory give $\beta^-/\alpha$ for Po$^{217}$. In a small sample of Em$^{221}$ practically free of Em$^{212}$ (obtained from a 100 Mev proton bombardment of thorium) it appeared that these activities were the same to within 5%. This would mean that the beta branching of Po$^{217}$ is less than 20%. This is not a sensitive measurement and the beta branching of Po$^{217}$ may be considerably less.

Attempts were made to collect Po$^{217}$ by alpha recoil from samples of Em$^{221}$. Recolls were collected in vacuum in the pulse analyzer chamber and the plate counted starting about 5 seconds after the end of collection. The results were negative. At equilibrium the Em$^{221}$ sample contained about 200 alpha counts per minute of Po$^{217}$. An upper limit of 10 seconds for the half-life of Po$^{217}$ is indicated if the assignment of the 6.54 Mev alpha particle is correct. This assumes 50 percent of the Po$^{217}$ atoms produced in the sample were collected on the recoil plate.
Fig. 14. Half-life of Em$^{221}$ from decay of At$^{217}$ (7.02 Mev) and Fr$^{221}$ (6.30 and 6.05 Mev) peaks on alpha pulse analyzer. Half-life of 6.54 Mev peak (Po$^{217}$) on alpha pulse analyzer.
The alpha particle energy of $\text{Em}^{221}$ is estimated to be $6.0 \pm 0.1$ Mev. Due to the low percentage of alpha emission in $\text{Em}^{221}$ and the high count from other activities in this region, this peak could not be detected.

The decay scheme indicated at present for $\text{Em}^{221}$ is shown in Figure 15.

The main difficulties encountered in these experiments were the interference from low mass emanation activities and the high beta-gamma activity from noble gas fission products which resulted in poor resolution on the pulse analyzer due to near-coincidences of beta and alpha pulses. The fact that fission product activities were troublesome here and not in previously mentioned studies indicates that the fission cross sections for lower atomic number noble gases are higher with respect to the cross section of $\text{Em}^{221}$ for 110 Mev protons than they are relative to the cross sections of low mass emanation isotopes in 340 Mev proton bombardments of thorium.

The amount of $\text{Em}^{212}$ relative to $\text{Em}^{221}$ increases rapidly with energy. Thus at 120 Mev $\text{Em}^{212}/\text{Em}^{221}$, on an atom basis, is 10, at 110 Mev it is 3, and at 100 Mev no $\text{Em}^{212}$ was observed in a sample containing only a few counts per minute of $\text{Em}^{221}$. These are rough values as the energy of the proton beam in the 184-inch cyclotron is only accurate to about 5 Mev at these energies. The problem is that the absolute yield of $\text{Em}^{221}$ is also dropping rapidly with energy, so that at 100 Mev not enough $\text{Em}^{221}$ is produced to be of use. The energy of 110 Mev used in most of the bombardments was a compromise between yield of $\text{Em}^{221}$ and freedom from neutron deficient emanation activities.
Decay scheme of Er-221.

- Fr-211
  - α, 8 min., β-
  - 6.30 Mev
  - Em 211

- At-217
  - α, 0.02 sec.
  - 7.02 Mev
  - Po-217
  - β-

- B-213
  - α, 47 min.
  - B-209
  - β-

- Po-213
  - 5.86 Mev
  - Pb-209
  - 3.3 hr.

- Pb-209
  - β-

Fig. 15
The difficulty presented by the high beta-gamma activity due to noble gases other than emanation was partially circumvented by a crude sort of fractional distillation. As these gases boil at much lower temperatures than emanation, it was possible to transfer them to another liquid nitrogen trap by simply removing the liquid nitrogen from the trap containing the sample for 20-30 seconds. The beta-gamma activity in the sample was high enough to register through the glass tubes on a survey meter, so that the transfer of the activity could be monitored and the distillation stopped before the emanation boiled over, but after most of the lighter gases were in the other trap. The sample remaining in the original trap was then transferred to the glow chamber and collected. Over 90% of the beta-gamma activity could be removed quickly and easily in this way without appreciable loss of emanation.

It was considered interesting, once the essential decay characteristics of $^{221}\text{Em}$ were known, to attempt to isolate it from the $4n + 1$ series ($^{233}$U decay series), where it is predicted to occur due to alpha branching in $^{225}\text{Ra}$. Using the methods of Perlman et al., the alpha particle energy of the most abundant group in $^{225}\text{Ra}$ is predicted to be about 4.8 Mev. The alpha half-life is probably in the range of $10^2 - 10^4$ years, the author being inclined to favor a prediction of the order of $10^3$ years. As the half-life of $^{225}\text{Ra}$ is 14.8 days this would indicate that an alpha branching ratio of $5 \times 10^{-4} - 5 \times 10^{-6}$ is predicted. W. Jentschke has published an estimate of less than $10^{-3}$ for the alpha branching of $^{225}\text{Ra}$.

A sample containing $2 \times 10^7$ disintegrations per minute of $^{229}\text{Th}$ was available in this laboratory through the courtesy of C. I. Browne, Jr.,
so attempts were made to isolate \( \text{Em}^{221} \) from this sample and collect it in the glow discharge for pulse analysis. The results indicated the presence of not more than two disintegrations per minute of \( \text{Em}^{221} \) at the time of separation from the \( \text{Th}^{229} \) solution. The yield in the experiment is not known, but \( 10^{-3} \) is probably a lower limit for the overall yield in the procedure. This would indicate an upper limit of \( 10^{-4} \) for the alpha branching of \( \text{Ra}^{225} \) (alpha half-life greater than 500 years). The above might yet prove to be a feasible method of obtaining sizeable pure samples of \( \text{Em}^{221} \) if larger samples of \( \text{Th}^{229} \) can be obtained or the efficiency of the collection process increased. In this case, it might also prove feasible to determine the alpha branching in \( \text{Ra}^{225} \) by using \( \text{Em}^{222} \) from known aliquots of \( \text{Ra}^{226} \) added to the \( \text{Th}^{229} \) solution to monitor the yield in the experiments.

In summary, \( \text{Em}^{221} \) is a 25 ± 2 minute activity decaying 80% by beta and 20% by alpha emission \((\beta^-/\alpha = 4 \pm 1)\). An alpha particle of energy \( 6.54 \pm 0.02 \) Mev was seen and observed to decay with a 25 minute half-life. It appears that this activity results from alpha branching in \( \text{Em}^{221} \) since it has the half-life of \( \text{Em}^{221} \) and does not belong to any of the beta decay products. On the basis of systematics, this activity is tentatively assigned to \( \text{Po}^{217} \), which appears to decay mostly by alpha emission with a half-life of ten seconds or less. The alpha particle of \( \text{Em}^{221} \) itself was not seen, but the data do not rule out its presence. As the \( \text{Fr}^{221} \) chain is well known, the identification of \( \text{Em}^{221} \) as a 25 minute beta emitter is considered well-founded.

7. Other Results for Emanation.---The preceding comprise all of the significant results for emanation obtained up to this time.
Actually, no serious effort was made to look for isotopes of emanation shorter-lived than $\text{Em}^{208}$ in samples produced by spallation. Present data indicate that no emanation isotope of mass less than $208$ with a half-life much greater than $10$ minutes and initial activity comparable to that of $\text{Em}^{212}$ is produced by spallation. Lighter emanation isotopes of odd mass number are expected to be difficult to detect because of their short electron capture half-lives, and in the case of $\text{Em}^{207}$, also because the expected alpha energy is the same as that of $\text{Em}^{208}$. It is possible that $\text{Em}^{206}$ has a half-life as long as $10$ minutes and appreciable alpha branching, so that its detection should be fairly easy. In one recoil sample from carbon bombardment of an enriched lead target ($\text{Pb}^{204} - 15\%, \text{Pb}^{206} - 50\%, \text{Pb}^{207} - 15\%, \text{Pb}^{208} - 20\%$) an alpha activity of energy $6.32 \pm 0.02$ Mev and roughly ten minute half-life was observed. This half-life and energy would fit those expected for $\text{Em}^{206}$ quite well. The opportunity did not arise to repeat this experiment, so these data are not given much weight at present. It is expected that carbon ion bombardments of lead will, however, prove to be the simplest way of looking for new emanation isotopes.

The cross sections for the production of emanations relative to the cross section of $\text{Em}^{212}$ were calculated for $340$ Mev proton bombardments of thorium. The calculated initial alpha activities at the end of a $50$ minute bombardment were: $\text{Em}^{212} - 11,000$ c/m, $\text{Em}^{211} - 105$ c/m, $\text{Em}^{210} - 2,100$ c/m, $\text{Em}^{209} - 1,200$ c/m, $\text{Em}^{208} - 2,380$ c/m, and $\text{Em}^{222} - 0.4$ c/m. The relative cross sections are calculated from these activities using the half-lives and branching ratios of the various isotopes to find the rate of formation of atoms of each in the bombardment. As the thorium foils could be considered thin targets to $340$ Mev protons, the
formula \( R = \sigma \eta n x \) holds, where \( R \) is the rate of production of atoms of a given nuclide (atoms per sec), \( \sigma \) the number of protons per second in the beam, \( \eta \) the cross section in cm\(^2\), \( n \) the number of target atoms per cm\(^3\), and \( x \) the thickness of the target in centimeters. Figure 16 is a plot of the relative cross sections calculated. A correction was made for the amount of Fr\(^{212} \) growing in from Fr\(^{212} \) after the end of bombardment and before removal of the emanation from the target on the assumption that the cross sections for Fr\(^{212} \) and Em\(^{212} \) are equal. As the correction was only 15% in this case, any reasonable error in the above assumption will not affect the results appreciably. The value for Em\(^{209} \) was calculated assuming \( EC/\alpha = 5 \) and the alpha activity of Em\(^{209} \) equal to 0.8 that of Em\(^{210} \) at the end of a 20-minute bombardment, thus it might easily be in error by 50%. The value plotted for Em\(^{208} \) is a lower limit, assuming 100% alpha decay. The value for Em\(^{222} \) should be regarded as somewhat of an upper limit as an unknown but perhaps appreciable amount of the activity observed could have resulted from the decay of Ra\(^{226} \) after the end of bombardment and before the removal of the emanation. It should be noted that the relative cross sections calculated in this way are not necessarily those for production of these isotopes by spallation, since these isotopes are not shielded. Their yields are thus the result of both spallation and the decay during the bombardment of their alpha and electron capture decay parents produced by spallation. A conclusion which appears to be justified, however, is that the total cross sections for the production of low mass emanation isotopes in these bombardments do not vary much over the range of mass numbers observed. This is often a useful bit of empirical information.
Fig. 16. Relative cross sections for emanation isotopes in 340 Mev proton bombardment of Th$^{232}$. 

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None of the numerous experiments performed yielded any positive evidence for low mass francium isotopes other than Fr$^{212}$. The results of the experiments in which francium was removed from thorium targets using the method developed by Hyde$^{18}$ indicate that no new low mass francium isotopes exist with half-lives greater than about five minutes and large alpha branching.

Experiments were performed in which emanation was separated from francium fractions isolated 5-10 minutes after bombardment in attempts to get information on electron capturing francium isotopes of appreciable half-life. Em$^{212}$, Em$^{210}$, and Em$^{211}$ were detected in such samples. Inspection of the results suggested, however, that most of the Em$^{211}$ and Em$^{210}$ was contamination resulting from emanation adsorbed on small amounts of solid residue from the target which were not completely removed by centrifuging. This conclusion was supported by the widely varying amounts of these activities relative to Em$^{212}$ found in the experiments and by the fact that the relative amounts of Em$^{210}$ and Em$^{211}$ were the same as those observed in emanation samples removed directly from the target. It thus appears that no new electron capturing francium isotopes exist with half-lives of greater than a few minutes. This result is not surprising. Fr$^{211}$ is the only undiscovered neutron deficient isotope of francium with an expected half-life of several minutes, and it is expected to decay mainly by alpha emission. Fr$^{210}$ and other isotopes of even mass number may decay largely by orbital electron capture, but are expected to have half-lives of a minute or less.
It appears unlikely that neutron deficient francium isotopes of mass other than 212 will be detected in thorium spallation products unless considerably faster chemical techniques can be developed which will yield pure samples of francium for alpha pulse analysis. The best hope for finding new low mass francium isotopes lies in carbon ion bombardments. At present facilities to enable chemical separation and pulse analysis of activities produced in such bombardments with half-lives much less than five minutes are not available, but there is hope that they soon will be. The gross alpha decay of bismuth targets bombarded by carbon ions indicated an activity of $0.5 \pm 0.1$ minute half-life produced in high abundance. This activity might be Fr$_{213}$ or Fr$_{211}$, or less probably Ac$_{214}$, but assignment of the activity and study of its decay characteristics must await future experiments.

To bring present francium data up to date, the results of an alpha ray spectrograph run on Fr$_{212}$ are reported below as communicated to the author by E. K. Hyde and F. Asaro (December 1952). Assuming the alpha particle energy of Em$_{212}$ to be exactly 6.262 Mev, the following alpha groups of Fr$_{212}$ were observed: 6.409 $\pm$ 0.007 Mev (37%), 6.387 $\pm$ 0.007 Mev (39%) and 6.339 $\pm$ 0.007 Mev (24%).

C. Radium

The presence of heavier radium isotopes and their daughters in spallation production of radium from thorium makes it impossible to pulse analyze such radium fractions directly in the hope of finding low mass, short-lived isotopes. Early evidence for the existence of an alpha emitting Ra$_{213}$ with a half-life of a few minutes has already been noted in the discussion of samples of Em$_{209}$ isolated from radium fractions.
Further information on Ra$_{213}$ had to await carbon ion bombardments. Several runs were made in which lead targets were bombarded for 5-10 minutes by carbon ions and then subjected to radiochemical procedures to remove radium. In three of these experiments an alpha particle of about 6.9 Mev energy and half-life of a few minutes was observed. In one run several hundred counts per minute of this activity were observed on the alpha pulse analyzer. The half-life of the activity was 2.7 ± 0.3 minutes and the energy of the alpha particle 6.90 ± 0.04 Mev. The decay curve obtained in this run is shown in Figure 17. After decay of the short-lived activity, several counts per minute of Em$_{209}$ were observed on the plate (identified by its alpha particle energy of 6.02 Mev and measured 30 minute half-life). No other activities were observed in significant amounts. The short-lived activity was identified as radium through the chemical separation, and the presence of Em$_{209}$ free of other emanation activities on the plate suffices to assign the activity to Ra$_{213}$ with a certainty equal to that of the assignment of Em$_{209}$. The alpha pulse analyses showed only the one peak assigned to Ra$_{213}$, but the possibility of appreciable electron capture is not ruled out as the alpha particle energy of Fr$_{213}$ may be close enough to that of Ra$_{213}$ that the two peaks were not resolved under the conditions of the pulse analysis. (See Figure 18).

Other low mass isotopes of radium with half-lives greater than one minute probably do not exist. As in the case of francium, the search for activities of such short half-life must await new facilities for carbon ion bombardments. Activities with half-lives as short as a minute or slightly less may eventually be detectable, so that there is at least a chance of finding Ra$_{212}$ in addition to getting a better determination of the properties of Ra$_{213}$. 
Fig. 17. Half-life of $^{213}$Ra from decay of 6.90 Mev peak on alpha pulse analyzer.
IV. DISCUSSION OF RESULTS

The discovery of Ra$^{213}$ fulfilled one of the primary purposes of these investigations, namely to show that the region of increased alpha stability around 126 neutrons exists for radium as well as for other elements of lower atomic number down to bismuth. This adds further to the empirical evidence for the 126-neutron closed shell.

The studies of the emanation isotopes of mass less than 212 have also established the shape of the alpha energy versus neutron number curve at and below 126 neutrons for this element. Figure 18 is a plot of alpha decay energy (alpha particle energy plus the recoil energy of the daughter nucleus) versus neutron number incorporating all present data on neutron deficient isotopes of elements from bismuth to radium. Data on heavier isotopes of bismuth, polonium and radium are also included to illustrate the sharp drop in alpha energy below 128 neutrons. The points for heavy isotopes of astatine, emanation and francium fall between the curves for polonium and radium and are omitted to avoid cluttering the figure. All data which did not result directly from these experiments were taken from the Table of Isotopes recently compiled at this laboratory. It will be noted that the shape of the curve for emanation at and below 126 neutrons is almost identical with the shapes of the astatine and polonium curves. On the assumption that this similarity holds for radium, francium and emanation isotopes as yet undiscovered, portions of the figure are filled in with broken lines to indicate estimated alpha decay energies. It might be remarked that the fact that the properties of the new emanation activities and Ra$^{213}$ fit the alpha systematics so well engenders considerable confidence in their present assignments.
Fig. 18. Alpha decay energy versus neutron number for isotopes of elements 83-88. For clarity, data on isotopes of elements 85-87 with neutron number greater than 126 are excluded.
It is noteworthy that the maximum alpha decay energy appears to occur in the 128 neutron isotopes. This is as expected as these decay into nuclides having 126 neutrons (the closed shell). The minimum alpha energy, however, occurs at 125 neutrons for polonium and emanation, and apparently for astatine, francium and radium as the 125 neutron isotopes seem to have the longest alpha half-lives for these elements. Alpha energies also increase rather slowly below 126 neutrons, the decay energy actually remaining less than that of the 126 neutron isotope for several lower neutron numbers. As alpha energies are a measure only of the difference in binding energy between the nuclide \((Z, N)\) and the nuclide \((Z-2, N-2)\), these observations in no way weaken the case for the 126 neutron closed shell, these effects being small at any rate with respect to the drop in decay energy from 128 to 126 neutrons. As \((Z, N)\) is more electron capture unstable than \((Z-2, N-2)\), addition of two neutrons per proton being required to achieve comparable beta stability in this region, the binding energy is normally decreasing more rapidly with decreasing \(N\) at \((Z, N)\) than at \((Z-2, N-2)\). This results in the usual increase in alpha decay energy with decreasing neutron number.\(^1\) The results indicate a lessening of this divergence in the binding energy curves for \(Z\) and \(Z-2\) just below 126 neutrons. This suggests that binding energies may decrease to an appreciable extent as a function of the number of neutrons removed from the closed shell just below \(N=126\). The minimum energy at \(N=125\) indicates that the binding energy of the 124th neutron in \(Z-2\) is considerably greater than that of the 126th neutron in element \(Z\). This fact and the relatively low decay energies of the next two lower odd \(N\) isotopes might be correlated with abnormally high pairing energies for neutrons just below the closed
shell, with a large increase in pairing energy of the 124th over the 126th neutron.

Certain comments on the results in the Em$^{221}$ experiments are in order, in particular as regarding the production of Em$^{212}$ by protons of 110 Mev energy on thorium. Closed cycle arguments and known and estimated decay energies yield the two equations:

$$\text{Pa}^{233} = \text{Em}^{212} + 5\alpha + n + 5\beta^- + 26 \pm 1 \text{ Mev}, \text{ and}$$

$$\text{Pa}^{233} = \text{Em}^{221} + 3\alpha + \beta^- + 16 \pm 1 \text{ Mev}.$$  

From these the energetic thresholds for the reactions $\text{Th}^{232}(p, 5\text{p16n})\text{Em}^{212}$, $\text{Th}^{232}(p, 3\text{p14n1a})\text{Em}^{212}$, $\text{Th}^{232}(p, \text{pl2n2a})\text{Em}^{212}$, and $\text{Th}^{232}(p, 5\text{p7n})\text{Em}^{221}$ are calculated to be 118, 90, 62 and 69 Mev respectively to within one Mev. These calculations assume that all the products have zero kinetic energy and are in the ground state. Conservation of momentum and the fact that protons emitted with zero kinetic energy have small chance of penetrating the coulombic barrier around the nucleus require that the actual threshold be higher than the energetic threshold by an unknown but probably considerable amount. Energies of the bombarding protons are accurate to within at least 5 Mev. It thus appears that the production of Em$^{212}$ from thorium by 110 Mev protons must involve the emission of alpha particles. The alpha particles need not, however, be emitted during the nuclear reaction itself. Thus Em$^{212}$ could be produced by either of the reactions, $\text{Th}^{232}(p, \text{pl2n2a})\text{Em}^{212}$ or $\text{Th}^{232}(p, \text{pl2n})\text{Th}^{220} \longrightarrow \text{Ra}^{216} \longrightarrow \text{Em}^{212}$. The energetic threshold of the first reaction is 62 Mev and that of the second is higher by the sum of the alpha decay energies of Th$^{220}$ and Ra$^{216}$ (est. 19 $\pm$ 1 Mev) or is about 80 Mev. Thus, the reactions $\text{Th}^{232}(p, \text{pl2n})\text{Th}^{220} \longrightarrow \text{Ra}^{216} \longrightarrow \text{Em}^{212}$ and $\text{Th}^{232}(p, 5\text{p7n})\text{Em}^{221}$, with
energetic thresholds of 80 and 70 Mev respectively, fit best the experimental observations which indicate that the actual threshold for the production of Em\textsuperscript{212} is slightly higher than that for Em\textsuperscript{221} and that cross sections are small in both cases, but relatively higher for Em\textsuperscript{212} at the proton energies used in bombardments. The experimental results indicate that there is little chance of obtaining sizeable samples of pure Em\textsuperscript{221} from proton bombardments of thorium. The isolation of emanation from Th\textsuperscript{229} and collection in the glow discharge may prove to be the best means to this end if large enough samples of Th\textsuperscript{229} can ever be obtained.

These investigations on the properties of isotopes of emanation, francium and radium can by no means be considered complete at this time. It is felt that carbon ion bombardments will in the future yield considerably more information on highly neutron deficient activities in this region. Similarly, much new information should be forthcoming from refined applications of the glow discharge collection technique to the study of emanation activities.
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