ALPHA ACTIVITY PRODUCED BY PROTONS OF MEDIUM ENERGY

A Thesis

Submitted in Partial Fulfillment of the Requirements

for the Degree of Doctor of Philosophy in the Faculty of

Graduate Studies and Research of McGill University.

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August 30th, 1951

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SUMMARY

Following the irradiation of thorium with 90 Mev. protons, two new alpha activities of 0.57 $\pm$ 0.15 seconds and 2.0 $\pm$ 0.3 seconds were observed. The 0.57 second activity has been assigned to a neutron deficient isotope of thorium, ${}_{90}Th^{224}$. The presence of this isotope had been previously established through its appearance in the U^{228} series. The half-life had not been measured.

On the basis of systematics, the 2.0 second period has been assigned to an isotope of protactinium ${}_{91}Pa^{225}$. This isotope is a member of a new alpha decay series, collateral to the 4n + 1 neptunium series. The alpha particle energies of the members of this chain have been measured. The series is given as

$$ {}_{91}Pa^{225} \xrightarrow{6.9} {}_{89}Ac^{221} \xrightarrow{7.6} {}_{87}Fr^{217} \xrightarrow{8.3} {}_{85}At^{213} \xrightarrow{9.2} {}_{83}Bi^{209} $$

The cross sections for the production of these isotopes by 90 Mev. protons have been estimated as

$$ \sigma\,(Th^{224}) \sim 1 \text{ milli-barn} $$

$$ \sigma\,(Pa^{225}) \sim 0.3 \text{ milli-barn.} $$

I. INTRODUCTION

General

During the past few years a large amount of information has become available concerning the elements in the region of the periodic table above lead. A sufficient number of new isotopes which decay by alpha particle emission have been produced to permit the regularities in behaviour to be tabulated. It was these regularities, as pointed out by various workers, that led to the present investigation into this region by the author. As a result of this research, an empirical plot has been made whereby the partial alpha half-life of an isotope decaying by alpha particle emission may be predicted. The conclusions to be drawn from this plot resulted in a search for protactinium mass number 225. This alpha emitting isotope has been produced by the proton beam in the McGill Synchrocyclotron, and its half-life measured as 2.0 ± 0.3 seconds, in good agreement with the predicted value. The assignment of this period to Pa^{225} has been made on the basis of the systematics of the alpha emitters. In addition, an alpha emitter with a half-life of 0.57 ± 0.15 seconds has been observed. This activity has been assigned to ${}_{90}Th^{224}$.

Historical

The study of alpha particle emission has been of great interest since it was first shown by Geiger and Nuttall[1] that there exists an empirical relation between the alpha particle range and the decay constant for the naturally occurring alpha emitters. This relation was expressed in the form

$$\log \lambda = a + b \log R \qquad \ldots\ldots\ldots\ldots\ldots(1)$$

where λ is the decay constant, R is the range of the alpha particle, b is a constant for the three naturally occurring series, and a is a constant for any one series but different for each of the three. This discovery remained an empirical fact until the advent of Quantum Mechanics provided the method whereby a satisfactory explanation for this relationship could be obtained(2,3,4). The application of a quantum mechanical treatment to the process of alpha decay yielded the expression

$$\log \lambda = \log \frac{v}{r} - \frac{2\pi zZe^2}{\hbar v} + \left(\frac{4e}{\hbar}\right)(2\ zZmr)^{\frac{1}{2}} \qquad \ldots\ldots\ldots\ldots.(2)$$

where λ is the decay constant

v is the relative velocity of the alpha particle

r is the radius of the nucleus formed in the disintegration

z is the charge of the alpha particle

Z is the charge of the daughter nucleus

e is the electronic charge

$\hbar$ is Planck's constant divided by 2π

m is the mass of the alpha particle.

This equation is equivalent to(1), provided the nuclear radius <u>r</u> remains constant. With the information available before 1939, this appeared to be the case.

Systematics

The completion of high energy particle accelerators has made possible the production of a great many new radioactive isotopes that decay by alpha particle emission. In the region above lead, more than seventy-five alpha active isotopes have been discovered, in addition to the natural decay chains. The availability of so much information in this neighbourhood has resulted in a study of the systematics of the alpha emitters by various authors[5,6,7,8,9,10,11].

The general trends have been shown up by these authors in a number of ways but probably the most useful is the method of Perlman[7].

In this case the alpha disintegration energy* is plotted against mass number, with the points of constant Z joined together. From this plot it is apparent that for any element, the lower the mass number, the greater the disintegration energy, until isotopes containing 127 neutrons are reached. At this point there is a sharp break, with the original trend reappearing to a certain extent in isotopes containing 126 neutrons or less. In the region above 127 neutrons, this representation results in a family of nearly parallel lines, with the feature that disintegration energies of as yet undiscovered isotopes may be estimated.

In addition to this method of indicating the general trends, alpha half-life versus alpha disintegration energy for the various classes of nuclei (even-even**, even-odd, odd-even, odd-odd) have been plotted. From these diagrams it is possible to observe definite regularities only for the even-even alpha emitters. The remaining classes indicate half-lives in excess of those predicted from the alpha disintegration energy on the basis of the Geiger-Nuttall relation. The factor by which the half-life exceeds the expected value shows no regularity for these three classes. In fact it is thought that in many cases the ground state alpha transition has not been observed, resulting in the departure factor being too low.

The usefulness of any of these plots is measured by the information that may be obtained from them concerning as yet undiscovered isotopes. Such quantities as half-life and alpha particle energy govern the techniques whereby an isotope is investigated. Accordingly, in the

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* Following the adopted practice, alpha disintegration energy is employed to denote the total energy of the ground state transition, including the energy of the recoil nucleus.

** This term refers to the number of protons and neutrons in the nucleus.

search for new isotopes, it is important to be able to predict within an order of magnitude the half-life, and to within a few million electron volts, the energy.

Predictions

In order to determine whether it is possible to predict alpha particle half-lives, a plot was made of the logarithm of the alpha half-life against the mass number. The value plotted here is the partial alpha half-life. This quantity is obtained by taking into account branching ratios where they are known to occur. In the region under consideration, the two competing modes of decay are alpha particle emission and orbital electron capture or beta emission. Assuming that there are two modes of decay, we have,

$$\frac{dN}{dt} = -\lambda N = -(\lambda_\alpha + \lambda_\beta)N$$

where λ_α and λ_β are the decay constants for alpha emission and orbital electron capture or beta emission respectively.

Now
$$\frac{1}{t} = \frac{1}{t_\alpha} + \frac{1}{t_\beta}$$

where t is the measured half-life of the decay and the subscripts refer to the two processes.

Also
$$\frac{\left(\frac{dN}{dt}\right)_\alpha}{\lambda_\alpha} = \frac{\left(\frac{dN}{dt}\right)_\beta}{\lambda_\beta}$$

therefore
$$t_\alpha = t\left[1 + \frac{\left(\frac{dN}{dt}\right)_\beta}{\left(\frac{dN}{dt}\right)_\alpha}\right] \qquad \text{............(3)}$$

In this expression the half-life t and the branching ratio $\frac{\left(\frac{dN}{dt}\right)_\beta}{\left(\frac{dN}{dt}\right)_\alpha}$ are known and hence the alpha half-life may be calculated. In many cases the branching ratios are not well known, but the estimates are probably good to within ten percent. The result of this plot is shown in Plate I. By joining points of the same mass type (even-even, etc.), for any element, it is seen that a family of curves is obtained. The general trends show up in the same manner as in the plots mentioned above, but in this case a fair estimate is available of the half-life.

Examination of this diagram resulted in the prediction that an isotope of protactinium, element number 91, containing 225 nucleons should decay by alpha particle emission with a half-life of from two to three seconds. Other predictions could be made but the decision to search for this isotope was governed by the availability of targets and the maximum energy of the proton beam in the McGill Synchrocyclotron*.

In estimating the possibility of forming this nucleus, use was made of the evaporation model of nuclear reactions(12). This model has been tested at lower energies in this region and found to be valid(13). The basic assumption is that the compound nucleus formed in a highly excited state, evaporates neutrons having an average energy of 2 or 3 Mev. The emission of a proton is also possible but because of the coulomb barrier, the probability is much less than for neutron emission. The number of neutrons evaporated from any nucleus with a given excitation energy depends upon the binding energy of the neutrons to that nucleus. In the case of thorium, the binding energy of the last neutron in Th^{232} has been measured as 6.0 ± 0.05 Mev.(14). Taking this value as being approximately the same for the binding energies of the neutrons to the compound nucleus $Th^{232} + p$, the threshold for evaporation of eight neutrons would be in the neighbourhood of 42 Mev., plus the difference

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* The maximum energy of the proton beam is 100 Mev. The term synchrocyclotron will be replaced hereafter by cyclotron.

between the proton and neutron binding energies. The fact that in the lower mass numbers the neutron binding energy will increase slightly would result in the threshold being higher by a few Mev. However, from this model it was possible to say that the reaction would certainly proceed at the proton energies available in the cyclotron.

It is important to note that all nuclei in this region are energetically unstable to alpha particle emission to a greater or lesser extent. The isotope ${}_{91}Pa^{225}$ decays by alpha emission to ${}_{89}Ac^{221}$, which is itself unstable, decaying to ${}_{87}Fr^{217}$ which in turn emits an alpha particle to reach ${}_{85}At^{213}$ which finally alpha decays to stable ${}_{83}Bi^{209}$. It can be said with little likelihood of experimental contradiction, that the half-lives of the daughters of ${}_{91}Pa^{225}$ will decrease with decreasing mass number and that the alpha particle energies will increase in the same manner. The half-life of any member of the chain will be less than its parent by approximately a factor of ten. As a result of this, there should be four alpha particle groups decaying with the half-life of the parent nucleus Pa^{225}. Of particular interest is the energy of the alpha particle emitted by ${}_{85}At^{213}$. This isotope of astatine has 128 neutrons and it is expected that the alpha particle emitted from this nucleus will be more energetic than any presently known, with the exception of the long range particles emitted in small percentages by RaC' and ThC'. The value for the alpha disintegration energy has been predicted[(8)] as 9.2 Mev. The alpha particle energy as measured in this experiment results in an alpha disintegration energy in good agreement with the expected value.

II. EXPERIMENTAL METHOD

General

The predicted value of two to three seconds for the half-life of $_{91}Pa^{225}$ indicated that use should be made of the existing rapid target extractor which has been described in detail previously[15,16]. This apparatus permits the study of isotopes whose half-lives extend down to about one-fifth of a second. The additional requirement to adapt this instrument to the measurement of the half-lives of alpha emitting isotopes was the construction of an alpha particle detector which would be insensitive to beta, gamma or X-rays. Such a detector was constructed.

In order to measure the energy of the alpha particles, a photographic plate technique employing nuclear emulsions was adopted. This method involved the registration of alpha tracks in type D-1 Ilford emulsions and the subsequent track counting with histogram representation. The considerations which led to the adoption of this method will be discussed below.

Rapid Target Extractor

In the measurement of activities whose half-lives are of the order of one second, it is not possible, with the exception of a few cases, to undertake a chemical separation. The target is mounted on a holder that runs on a track inside a stainless steel tube which is inserted into the vacuum chamber through the probe gate. The back end of the holder is attached to a rod which runs through a vacuum seal to a piston chamber. By the application of compressed air to the piston, the target may be withdrawn from the bombardment position to the counting position, which is still in the main vacuum, in a time of the order of one-fifth of a second. At the counting position there is a housing into which the counter assembly is inserted. A vacuum seal is maintained by this assembly at the housing so that all counting is carried out without disturbing the vacuum system of the cyclotron.

The recording of half-lives is carried out at a location in the tunnel well removed from the cyclotron itself. The signal is fed up a coaxial line and into an Atomic Instruments Linear Amplifier. The discriminator output is coupled to a counting rate meter and to a Berkeley Model 2000 scaler. The output from the counting rate meter is applied to the horizontal deflecting plates of a Dumont 241 oscilloscope fitted with a Fairchild Oscillo-Record camera. A selected linear sweep is applied to the vertical plates and brightening pulses at a predetermined repetition rate are applied to the z-axis amplifier. This display is photographed during the decay and is analysed to obtain the half-life. In order to provide an abcissa, a base line is photographed on the same frame prior to each exposure. At the same time that the output of the counting rate meter is being photographed, the scaler is in operation for a predetermined length of time, as a rule for a few half-lives. Following the display on the oscilloscope, which usually covers four or five half-lives, the background counting rate is recorded from the scaler. The cycle of operation lasts a few minutes, and then may be repeated. A block diagram of the apparatus is shown in Figure 1.

In dealing with short lived activities, it is convenient to radiate the target only for a period equal to one or two half-lives. If there are longer lived activities present, longer bombardments will only serve to increase the amount of background. Precise and reproducible radiations are obtained with the aid of the cyclotron oscillator pulser(17). The pulsed oscillator is set in operation and after a desired length of time, is automatically shut off. At the same time, the target is extracted to the counting position. When it arrives there, the recording of the decay as described above is commenced. After initiating the pulser, all operations are automatic until the background is measured on the scaler. This arrangement removes the element of human error as nearly as possible from the operation, and allows for reproducibility which is important when comparative results are required.

Alpha Particle Detector

The two main features that should be incorporated into a detector to register alpha particles are low sensitivity to other radiations and good counting geometry with respect to the target. Both these requirements have been met with the use of a zinc sulphide screen viewed through a short lucite light piper by a photomultiplier tube.

This arrangement has several advantages over the use of a proportional counter. Since the sources employed for the half-life measurements were infinitely thick for alpha particles, the distribution in energies would be from zero up to the maximum. In order to take advantage of as many disintegrations as possible, there should be the minimum amount of absorbing material between the source and the detector. In the case of the zinc sulphide detector, the only requirement is a light shield to protect the photomultiplier tube. A proportional counter on the other hand, requires a window which is much thicker, thus cutting down on the efficiency of detection. Another consideration is that since the target remains in the vacuum throughout, the detector must also extend onto the vacuum system. In order to maintain good counting geometry with a proportional counter, i.e. large window, it would be necessary to employ a thick window to safeguard the vacuum. This undesirable feature is removed by the present arrangement in which the lucite rod acts as the vacuum seal.

The type of photomultiplier employed is the RCA 5819. This tube has a large sensitive area which allows good counting geometry without the necessity of employing a truncated cone. The spectral response of the 5819 peaks at 4800 Å with a range of maximum value given by the manufacturers from 4300 Å to 5300 Å. In choosing the type of zinc sulphide to be employed, it is desirable to match as nearly as possible the response of the phototube. The two types of zinc sulphide available for use as scintillation detectors are zinc sulphide activated with copper, having an emission wavelength of 5200 Å [18] and silver activated zinc sulphide whose emission wavelength is 4500 Å. RCA 33-Z-20A ZnS:Ag has been employed since its emission time is considerably less.

The zinc sulphide screen is prepared by transferring the adhesive part of scotch tape to a thin sheet of mica. This is accomplished by placing scotch tape in a solution of petroleum ether for a few seconds and then removing the adhesive material which dissolves to a certain extent in this solvent. After spreading a uniform layer over the mica, the ZnS:Ag is powdered on with a fine nylon brush and the excess tapped off. The adhering layer is very firmly held and appears to be quite uniform. This method is fairly reproducible, giving rise to a thickness of approximately 3.8 mg/cm^2.

The lucite light piper is surrounded with aluminum foil to act as a reflector, outside of which is wrapped scotch tape with the adhesive outwards. The mica screen is placed on one end of the lucite rod and covered with five layers of beaten aluminum foil, the total thickness being 0.9 mg/cm^2. This foil is held in place by the scotch tape and the whole nose is light tight. The opposite end of the rod is flared, and rests against an O-ring to provide a vacuum seal. The surface of this end is shaped as nearly as possible to fit the glass envelope at the photo sensitive surface of the 5819.

The effect of the magnetic field of the cyclotron on the operation of the phototube is overcome by means of magnetic shielding. Two concentric cylinders of soft iron tubing are placed around the tube in addition to the μ -metal shield that is used under normal circumstances. This arrangement forms a vacuum tight unit which is inserted into the housing of the rapid target extractor.

The preamplifier circuit is built into the back end of the shielding unit. The output pulse from the phototube is inverted and passed through a cathode follower which feeds the one hundred feet of coaxial line between the cyclotron and the recording units. The circuits are based on those described elsewhere(19). The time constants of the input and output of the inverter have been made equal to the decay constant of the phosphor, as has been suggested previously(20). The entire unit is shown in Plate II.

The unit was tested to determine the effect of beta and gamma background on the operation of the counter. In the presence of up to 15 r/hr. of ionizing radiation originating from beta and gamma rays, there was no detectable count above ordinary tube noise. At the same time, the ability of the counter to detect alpha particles was not impaired.

Counting Losses

Since the decay time of the phosphor has been variously reported[21,22] as 2-10 micro-seconds, the effect of resolving time on the recorded counting rate becomes important.

The true counting rate N as a function of the recorded counting rate R may be expressed in the form of a power series[23],

$$N = R + \tau R^2 + \nu R^3 + \text{----}$$

The parameters τ and ν must be evaluated in order to determine the true counting rate.

In the case where the counting losses are not considerable, i.e. roughly less than twenty percent, the second correction term may be neglected. The first approximation is then

$$N = R + \tau R^2.$$

The actual value of τ may be obtained in the following way. Two sources of approximately the same strength are counted separately and together. If the recorded counting rates per second are R_a, R_b and R_{a+b}, then it follows that

$$\tau = \frac{R_a + R_b - R_{a+b} - b}{R_{a+b}^2 - R_a^2 - R_b^2} \quad \text{sec/count} \quad \ldots\ldots\ldots\ldots (4)$$

where b is the background counting rate.

The values obtained for τ at different counting rates encompassing those encountered in practice are listed in Table 1.

Table 1

R_a c/sec.	R_b c/sec.	R_{a+b} c/sec.	τ sec/c.
8506	3591	11328	17.9×10^{-6}
6031	2961	8520	17.3×10^{-6}
996	498	1481	17.1×10^{-6}
5867	5595	10874	15.1×10^{-6}

The average value obtained for τ is $16.9 \pm 1.22 \times 10^{-6}$.

Nuclear Emulsion Technique

The foremost consideration of any technique employed to obtain alpha particle energies is that the source be thin. In cases where chemistry is performed after bombardment, this condition is readily fulfilled. In the present case, however, a chemical separation is not carried out, with the result that the target must also serve as a source. Any source that is thin for alpha particles of the order of ten million electron volts, is extremely thin to protons of from 50 to 100 Mev. Under these circumstances the number of collisions made by the proton beam will be small, resulting in a very low counting rate. As was pointed out above, the measured half-life is that of an alpha emitting parent which decays through three alpha emitting daughters to stable bismuth. It is the energies of these four alpha particles that are to be measured, and hence there will be a factor of four less than the number available for half-life measurements.

The standard method of measuring alpha particle energies is by the use of a proportional counter or ionization chamber. Either of these arrangements, coupled with a channel analyser, would give satisfactory results under normal circumstances. In the case of low counting rates such as those encountered in this experiment, the poor geometry resulting after collimation of the alpha particles would leave too few counts per channel to be of any significance. In the case of short half-lives, the use of a single channel analyser requires repeated bombardments with the resulting increase in background from the other alpha emitters present.

The use of ZnS:Ag as a means of measuring alpha particle energies has been investigated by several workers[21,22,24], and the author. In all reported cases the spread in pulse height of a monochromatic alpha beam was found to be too large to be of any use. This spread in pulse height has been attributed to scattering of the light in the powder.

In the nuclear emulsion technique employed in this experiment, no attempt was made to measure half-lives in conjunction with alpha particle energies. The target was bombarded for a short period and then rapidly placed next an Ilford type D-1 nuclear emulsion. Following a short exposure to the photographic plate, the target was removed so that no further alpha particles could register in the emulsion. This practice was adopted to reduce to a minimum the tracks caused by longer lived alpha activities which are present as a background. The tracks recorded during the exposure were measured and the equivalent energies calculated, making use of the range-energy relation for alpha particles in Ilford emulsions as determined by Lattes et al[25]. A histogram plot was then made to show the alpha particle groups of the new series.

Since the nuclear plate must remain in the vacuum chamber during the bombardment period, it was necessary to shield it very thoroughly from scattered protons. At the same time a channel had to be available so that the target could be placed next the emulsion directly the radiation was completed. The arrangement adopted and found suitable is shown in Plate III.

The nuclear emulsion was enclosed in a container with walls one-half inch thick, which is sufficient to stop protons of 95 Mev.[26]. The channel was in the form of an arc of a circle, the entrance to which was shielded from the protons scattered directly from the target. The target itself was held on one end of a piece of thin polystyrene, bent in the form of an arc with the same radius of curvature as that of the chennel. The other end of the polystyrene was attached to an arm which permitted the target to be rotated through nearly 180° from the bombarding position up the channel to the nuclear emulsion. The rotation of the target was accomplished by means of a flip coil connected to the shaft that held the arm. By energizing this coil, the target could be swung around to the nuclear plate and by reversing the current in the coil, returned to the original position.

This arrangement was satisfactory in that the scattered protons did not reach the nuclear plate. However, the small angle scattering from the target resulted in a considerable portion of the beam striking the copper shielding after circling the centre of the cyclotron. The energy of the protons was sufficient to cause reactions of the type (p,xn) in the copper, with the result that some knock-on protons were recorded in the emulsions. Since the track of a proton of two Mev. could be confused with that of an alpha particle of about eight Mev., it was highly desirable that the distinction between the two types of track be definite. The types of emulsion tested were

C-2, E-1, and D-1. Of these, the D-1 contains the finest grain and is recommended by the manufacturers for the recording of fission fragments. Only in this type of plate was it possible to distinguish definitely between knock-on proton tracks and alpha particle tracks. In spite of the fact that distinction could be made directly in a majority of the cases, since the alpha tracks commence at the surface, and most of the knock-on proton tracks occur within the emulsion, it was considered advisable to take as many precautions as possible. The use of D-1 emulsions resulted in a much cleaner plate which facilitated the track counting.

Target Preparation

The targets employed for the half-life determinations required that the maximum density of thorium itself be present. Since thorium metal was not available, the targets were constructed using thorium oxide. The powder form in which this material exists made it necessary to employ a rigid backing to which the thorium oxide could be attached. The first procedure followed was the fusion of ThO_2 on a nickel foil with the aid of $KHSO_4$. This was superceded when it was discovered that the counting rate could be increased considerably by applying the ThO_2 to the backing material with the aid of the adhesive part of scotch tape, as was described above in the construction of the scintillation screen. The backing material employed in these cases was silver.

In the case where the energies of the alpha particles were to be measured, the procedure followed was the one reported by Kahn[27]. A solution containing $Th(NO_3)_4$ and a small amount of $K_3Fe(CN)_6$ was electrolysed. The deposit on the cathode which consisted of $ThFe(CN)_6$, was ignited to the oxides of thorium and iron and the latter leached out with 12N HCl, leaving the ThO_2 on the cathode. The amount of ThO_2 deposited was estimated from the results of Kahn to be in the neighbourhood of 500 micro-grams per square centimeter.

The energy loss of an alpha particle traversing this thickness can be estimated with the aid of the empirical rule formulated by Bragg and Kleeman[28]. This rule states that the stopping power of an atom is proportional to the square root of the atomic weight. In order to compare the range in the substance with the range in air, the rule is placed in the form

$$\frac{R_a d_a}{R_s d_s} = \frac{\psi_a}{\psi_s} \qquad \text{..................(5)}$$

where R is the range, d is the density, and ψ is the permeability of the substance to alpha particles. The subscripts a and s refer to air and the substance respectively. Comparison of the calculated range with values given by W. A. Aron[26] indicate that an eight Mev. alpha particle would lose approximately 80 Kev. in traversing a thickness of $500 \mu gms/cm^2$ of thorium oxide.

That this method may be open to large errors has been pointed out by Wu[29]. The fact that a known weight of a material has been deposited is no guarantee that it is distributed evenly over the surface. This observation has been confirmed, as in some cases the alpha particle spectrum was flat with a tail extending down to 4 Mev.

III. RESULTS

General

The bombardment of thorium oxide produced two short-lived alpha activities. The half-lives of these isotopes have been determined as 0.57 ± 0.15 sec. and 2.0 ± 0.3 sec. The relative yield of the two activities was such that the periods could not be separated directly from one decay curve. The process through which they were resolved will be described below.

The periods have been assigned as follows. The 0.57 sec. activity is presumed to belong to an isotope of thorium, Th^{224}, and the 2.0 sec. activity to an isotope of protactinium, Pa^{225}. The thorium isotope is a member of an already established series[30], but the half-life had not been determined. The protactinium isotope is a member of a new 4n + 1 series. The alpha particle energies of this decay chain have been measured during this investigation.

Half-life determinations

The results of the first series of bombardments are given in Table II. All information pertinent to the succeeding discussion is listed in this and the following tables. The target in this case consisted of ThO_2 fused onto a nickel foil.

Table II

Run No.	Bombarding time secs.	Proton energy Mev.	Rate meter time const. milli-secs.	Time base speed secs.	Time pips no/sec.	Half-life secs.	Initial counting rate c/sec.
Th-1-3	2	80	41	5	2	1.3_0	1140
4	2	80	41	5	2	1.2_2	2170
6	2	90	103	5	2	1.1_5	3600
7	3	90	103	5	2	1.2_4	3900
8	10	90	103	10	2	1.7_1	1500
10	3	90	103	7	2	1.3_6	722
11	3	90	103	7	2	1.4_1	2500
12	3	90	103	7	2	1.4_6	2640
13	3	90	103	7	2	1.4_4	2510
14	3	90	103	7	2	1.4_1	1840

From the above values it appeared that, with the exception of number Th-1-8, a half-life of 1.33 ± 0.08 seconds was present. The fact that Th-1-8 indicated a longer half-life, at this time was attributed to the longer bombardment period and that the large amount of background activity present resulted in considerable uncertainty in plotting the decay curve. In addition, time marker pips at the rate of two per second were unsuitable, providing too few points for the curve on the shorter time bases. At this time, marker pips were available at the rate of two, twenty, and two hundred per second, the latter two rates being too high for a half-life of this value.

In order to obtain a more precise value for the half-life, a scale of two was employed to reduce the twenty marker pips per second to ten. The results of this new series are given in Table III.

Table III

Run No.	Bombarding time secs.	Proton energy Mev.	Rate meter time const. milli-secs.	Time base speed secs.	Time pips no/sec.	Half-life secs.	Initial Counting Rate c/sec.
Th-3-3	2	90	103	5.4	10.08	0.9_7	7110
5	2	90	41	5.4	10.08	1.0_7	12800
7	1.5	90	41	5.4	10.08	1.1_2	4860
9	1.5	90	41	5.4	10.08	1.0_0	3150
10	1.5	90	41	5.4	10.08	1.1_1	2570

The values of the half-life obtained from this radiation did not correspond to those listed in Table II. The type of target employed in this second run consisted of thorium oxide powder mounted on a silver backing. Since the activity was increased by a factor of at least two, it was felt that these values might be more representative of the true half-life.

Accordingly, a third radiation was carried out. The conditions of this bombardment and the resulting periods are listed in Table IV.

Table IV

Run No.	Bombarding time secs.	Proton energy Mev.	Rate meter time const. milli-secs.	Time base speed secs.	Time pips no/sec.	Half-life secs.	Initial Counting Rate c/sec.
Th-4-17	1.5	76	103	5	10.01	1.1_9	620
31	1.5	80	41	3	10.01	0.7_5	1660
33	1.5	80	103	3	10.01	1.0_4	1790
37	3	80	258	8	2	2.2_0	1240
38	3	80	258	8	2	1.7_4	735
44	3	80	103	10	2	1.6_6	1040
21	1.5	85	103	5	10.01	1.2_5	1500
22	1.5	85	103	5	10.01	1.2_5	2030
4	1.5	90	103	6	10.01	1.1_3	2080
5	1.5	90	103	3	10.01	0.8_5	2720
6	1.5	90	41	3	10.01	0.9_6	4740
8	1.5	90	41	3	10.01	0.9_8	2700
9	1.5	90	41	3	10.01	0.8_8	3790
23	1.5	90	103	5	10.01	1.2_8	2030
24	1.5	90	103	5	10.01	1.0_8	2010
25	1.5	95	103	5	10.01	0.9_8	1700
26	1.5	95	103	5	10.01	1.0_7	1310
27	1.5	95	103	3	10.01	1.0_9	2630
28	1.5	95	41	3	10.01	0.9_7	2970
29	1.5	95	41	3	10.01	0.9_5	3200

The spread in values obtained from this run was more than could be expected on the basis of statistics alone. A critical survey of the results indicated that although the effect might not appear large, a longer integrating time constant in the counting rate meter resulted in a slightly longer value for the half-life. In addition to this, comparison with the results listed in Table III showed that other conditions being equal, a longer time base resulted in a longer measured half-life.

In the presence of only one decaying period, the value for the half-life should not be effected noticeably by varying the time constant of the counting rate meter, provided it is shorter by at least a factor of ten than the period of the activity being measured. The treatment of the problem of the response of the counting rate meter with an RC time constant t_2 to impulses originating from an exponential decay with a time constant t_1, is identical with the solution of parent and daughter activities with half-lives t_1 and t_2 respectively. In the presence of two decaying activities, the use of a time constant in the counting rate meter that is too long will tend to suppress the shorter activity.

During the cycle of operation in the measurement of a half-life with the rapid target extractor, there is a time delay at the commencement of the recording, equal in length to one-sixth of the time base. This delay is constant, depending only upon the rate of rotation of a cam which governs the length of the time base. In the case of two isotopes decaying with short periods, the value obtained for the apparent half-life would depend upon the time interval at which it was measured.

As a consequence of these considerations, it appeared that the activity measured was, in fact, a composite decay, with the values for the half-lives too close together to permit them to be resolved directly on the decay plot. The possibility was present of considerably enhancing one period over the other by varying the bombarding time and the time base interval. This had, in fact, been demonstrated by radiations Th-4-37, 38 and 39, the measured values for the decay being much longer in these cases. It should be pointed out that in spite of the long bombardment, the initial activity was not great.

In order to verify that the measured half-life did depend upon bombarding time and recording conditions, the radiations listed in Table V were carried out.

Table V

Run No.	Bombarding time secs.	Proton energy Mev.	Rate meter time const. milli-secs.	Time base speed secs.	Time pips no/sec.	Half-life secs.	Initial Counting rate c/sec.
Th-5-2	0.6	90	16	2	20.00	0.8_1	1620
9	3	90	41	8.2	1.83	1.4_3	1550
10	3	90	41	8.2	1.83	1.2_9	1780
12	3	90	41	8.2	1.83	1.3_6	1750
13	3	90	41	8.2	1.83	1.2_6	1840
17	3	90	41	8.2	1.83	1.8_6	1940
Th-9-4	0.6	90	41	2.35	20.00	0.8_6	1520
5	0.6	90	41	2.35	20.00	0.8_5	1370
8	0.6	90	41	2.35	20.00	0.8_8	1150
9	0.6	90	41	2.35	20.00	0.7_4	1420
10	0.6	90	41	2.35	20.00	0.8_8	1250
11	0.6	90	41	2.35	20.00	0.8_0	1280
12	0.6	90	41	2.35	20.00	0.8_5	1470
13	0.6	90	41	2.35	20.00	0.7_9	1410

The range of values agreed fairly well when the bombarding time and recording conditions remained constant, but the change in these conditions produced a considerably different set of measurements for the half-life. The only conclusion to be drawn from this behaviour was that a mixture of activities was present, as was indicated to some extent previously. Approximate values for the half-lives of the separate activities may be obtained as follows.

In the case where two activities are present,

$$\frac{dN}{dt} = -\lambda_1 N_1 - \lambda_2 N_2$$

where $\frac{dN}{dt}$ is the counting rate, λ the decay constant, and N the number of radioactive atoms present. The subscripts refer to the two activities. Now

$$N = N_0 e^{-\lambda t},$$

where N_0 is the number of radioactive atoms present at time $t = 0$.

The time $t = 0$ is taken at the end of bombardment, and at that time

$$N_0 = S(1 - e^{-\lambda T}),$$

where T is the radiation time and S is the number of radioactive atoms present when T becomes infinite, i.e. the number of atoms at saturation. Thus,

$$\frac{dN}{dt} = -\lambda_1 S_1 e^{-\lambda_1 t}(1 - e^{-\lambda_1 T}) - \lambda_2 S_2 e^{-\lambda_2 t}(1 - e^{-\lambda_2 T})$$

Also,

$$\frac{dN}{dt} = -\lambda_m(N_1 + N_2),$$

where λ_m is the measured value of the decay constant. In the case of two activities that are not resolved on the decay curve, the value of λ_m may be considered to remain constant over approximately one measured half-life.

Hence,

$$\lambda_m(N_1 + N_2) = \lambda_1 S_1 e^{-\lambda_1 t}(1 - e^{-\lambda_1 t}) + \lambda_2 S_2 e^{-\lambda_2 t}(1 - e^{-\lambda_2 T}),$$

or

$$S_1 e^{-\lambda_1 t}(1 - e^{-\lambda_1 T})(\lambda_1 - \lambda_m) = S_2 e^{-\lambda_2 t}(1 - e^{-\lambda_2 T})(\lambda_m - \lambda_2) \qquad \ldots\ldots\ldots\ldots\ldots(6)$$

from which

$$\ln S_1 - \ln S_2 - \lambda_1 t + \lambda_2 t + \ln\left(\frac{1 - e^{-\lambda_1 T}}{1 - e^{-\lambda_2 T}}\right) + \ln\left(\frac{\lambda_1 - \lambda_m}{\lambda_m - \lambda_2}\right) = 0 \quad \ldots.(7)$$

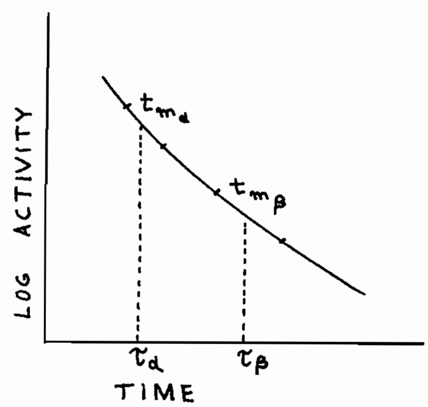
Consider the two cases in which the period of bombardment is the same but the time after the end of the radiation when the value for the measured half-life is taken, is different. There will be two equations (7) involving λ_1, λ_2, λ_{m_α}, $\lambda_{m\beta}$, τ_α, and τ_β, where the subscripts α and β refer to the two conditions. From these two equations, one equation may be obtained.

$$\ln\left[\frac{t_{m\alpha} - t_1}{t_{m\beta} - t_1} \cdot \frac{t_2 - t_{m\beta}}{t_2 - t_{m\alpha}}\right] = 0.693 \left(\frac{t_1 - t_2}{t_1 t_2}\right) (\tau_\beta - \tau_\alpha) \ldots\ldots.(8)$$

In this equation, $t_{m\alpha}$ and $t_{m\beta}$ are the measured half-lives at τ_α and τ_β respectively. Thus, if t_1 is known, t_2 may be evaluated.

The gross decay is shown schematically in the accompanying diagram. The values of τ_α and τ_β are obtained by employing two different time bases, and the actual time is given by

$$\tau = \frac{1}{6}\text{ time base} + \frac{t_m}{2}$$



The assumption made is that the slope of the line joining two points on the composite decay curve one half-life apart is approximately equal to the tangent drawn to the curve at the mid point.

If all measurements are carried out at the same bombarding energy, the ratio of the cross sections will remain constant, and hence

$$\frac{S_1}{S_2} = \text{constant} = k$$

Thus from (6)

$$k = \frac{e^{-\lambda_2 \tau}(1 - e^{-\lambda_2 T})\,(\lambda_m - \lambda_2)}{e^{-\lambda_1 \tau}(1 - e^{-\lambda_1 T})\,(\lambda_1 - \lambda_m)} \quad \text{..................(9)}$$

Employing the values of λ_1 and λ_2 obtained in the solution above, the constant k may be evaluated with the use of a different set of values for τ, T, and λ_m.

It is then possible to check this value of k with a third new set of values for τ, T, and λ_m.

In Table VI are listed the values of the parameters employed in the solution of the three equations, together with the bombardments from which they are taken.

Table VI

Equation number	t_{m_1} sec.	t_{m_2} sec.	t_m sec.	T sec.	τ sec.	$\tau_2 - \tau_1$ sec.	Reference bombardment
8	0.9_4	1.0_6	—	1.5	—	0.4_6	Th-3-7,9 Th-4-6,8,9
9	—	—	1.3_4	3.0	2.0_6	—	Th-5-9,10,12,13.
9	—	—	0.8_3	0.6	0.8_2	—	Th-9-4,5,8,9,10,11,12,13.

Since the equations to be solved are transcendental, no direct solution is possible. The method of solution involves a trial solution with a t_2 and t_1 obtained from equation (8). The final values for these periods are derived by successive approximation until all conditions are satisfied. The values obtained by this method are

$$t_1 = 0.57 \pm 0.15 \text{ sec.} \quad \text{................(10)}$$

$$t_2 = 2.0 \pm 0.3 \text{ sec.} \quad \text{................(11)}$$

Four typical photographs obtained from the oscilloscope display are shown in Plates IV, V, VI and VII.

Production Cross Sections

The ratio of the cross sections for the production of the two isotopes at a bombarding energy of 90 Mev. may be calculated with the aid of equation (9).

The saturation activity is produced when the number of radioactive atoms formed in unit time is equal to the number of disintegrations per unit time. The number of radioactive atoms produced is

$$n_p \sigma N,$$

where n_p is the number of protons impinging per second on a target containing N atoms per cm^2. and σ is the cross section for the reaction. The number of atoms decaying per second is λS, where λ is the decay constant and S is the number of radioactive atoms present at saturation. Hence

$$n_p \sigma N = \lambda S.$$

In the present case,

$$n_p \sigma_1 N = \lambda_1 S_1$$

and

$$n_p \sigma_2 N = \lambda_2 S_2.$$

Therefore

$$\frac{\lambda_1 S_1}{\lambda_2 S_2} = \frac{\sigma_1}{\sigma_2} \qquad \text{..................(12)}$$

The value of the ratio $\frac{S_1}{S_2}$ is determined from equation (9) above, and since the half-lives are also known, the ratio of the cross section is obtained.

The value of k was 1.0_0, and thus the ratio of the cross sections is given by

$$\frac{\sigma_1}{\sigma_2} = \frac{t_2}{t_1} \times 1.0_0 = 3.5 \pm 1 \qquad \text{...........(13)}$$

From the above results it is possible to place a lower limit on the absolute cross sections for the production of these isotopes. The cross section σ is given by

$$\sigma = \frac{\text{Number of transformations}}{\text{Number of incident protons x number of atoms present.}}$$

The sources employed were infinitely thick for alpha particles and thus all particles emerging from a depth equal to the range in the source would be recorded, provided the detector was unshielded. The aluminum foil covering the ZnS screen was 0.9 mg/cm^2, which is sufficient to stop alpha particles of 1 Mev. The alpha particles must leave the source with at least this energy in order to reach the screen.

The range of an eight Mev. alpha particle in ThO_2 is approximately 34 mg/cm^2. Assuming that the effective source thickness is 30 mg/cm^2 and the beam current at the outer radii is of the order of 1/20th μamp, it is possible to calculate the cross section. The cross section calculated on the basis of the initial counting rate will be too high by a factor of four, owing to the fact that there are three daughter activities which will be incorporated in the total activity. Due to the fact that the recording system employs a discriminator, which is adjusted to eliminate amplifier and photo-tube noise, a number of counts will be lost. There is no manner in which the number of counts thus lost may be estimated.

The estimated values for the production of the isotopes is

$$\sigma(2.0 \text{ sec.}) = 0.3 \text{ milli-barns}$$

$$\sigma(0.57 \text{ sec.}) = 1.0 \text{ milli-barns.}$$

No claim is made on these values other than that they are probably within a factor of ten of the actual cross sections.

Yield Curve

The yield curve for the composite activity was determined. The relative yield at different bombarding energies is given in Table VII.

The resulting curve is shown in Figure 2. The corrected yield is obtained by taking into consideration the fall off in proton beam current with increasing radius. This variation of beam current with energy of the proton beam is shown in Figure 3.

Table VII

Proton bombarding energy Mev.	Relative yield uncorrected arbitrary units	Relative yield corrected arbitrary units
63	55	55
67.5	136	166
71.5	130	487
76	384	534
80	558	798
85	1117	1700
90	1170	2150
95	875	3040

The energy of the bombarding protons was determined by the radial position of the target, rather than by direct measurement. The internal beam of the cyclotron has been studied by W. H. Henry[31], who has found that radial oscillations exist which cause the beam to be inhomogeneous in energy. The spread in proton energies at the radii employed in this experiment has been estimated by Henry to be of the order of ten percent. This condition prevents any definite conclusions to be drawn from the yield curve other than that the reaction progresses more favourably as the energy of the incident protons is increased.

Alpha Particle Energies

The histogram representation of the measured alpha particle energies is shown in Figure 4. A target prepared as described previously was bombarded for a period of three seconds and then placed near the nuclear emulsion for a similar length of time. The resulting tracks were counted and segregated into groups according to energy. The relation employed to convert length of track into alpha particle energy was that given by Yagoda[32], based on the results of Lattes et al[25].

In order to determine the resolution that might be expected, a test was carried out using Pb^{212} collected from Th^{228} as a standard source. From this calibration plate, which is shown in Figure 6, it was apparent that the resolution was at best of the order of five percent.

The fact that there are not four equal peaks in the distribution indicates that there is a mixture of alpha particle groups present. At the high energy end of the histogram there is a peak at 9.2 Mev. This alpha particle is assigned by systematics to ${}_{85}At^{213}$. The fact that At^{213} is the fourth of a series of alpha emitters originating with ${}_{91}Pa^{225}$, indicates that this isotope of protactinium has been produced, and also serves as a guide in determining the ratio of the Pa^{225} series to the other alpha particle groups present in the histogram.

The background activity produced in a three second bombardment was monitored separately for periods and relative abundance. A number of long half-lives were produced and it was not possible to separate them individually. The total number of disintegrations that took place in a three second period directly following the bombardment was of the order of 5% of the number of disintegrations due to the two short activities.

IV. INTERPRETATION OF RESULTS AND DISCUSSION

The results of the proton bombardment of thorium at 90 Mev. indicate the presence of two new half-lives, namely,

1. $t_{\frac{1}{2}} = 0.57 \pm 0.15$ secs.
2. $t_{\frac{1}{2}} = 2.0 \pm 0.3$ secs.

In order to make the correct assignment of these activities, it is necessary to consider:-

1. The systematics of the alpha emitters in the region above lead.
2. Thresholds and cross sections.
3. The spectrum of the emitted alpha particles as read from the histogram.

1. It was pointed out in the introduction that the systematics of the even-even alpha emitters are the most consistent. In view of this, it might be expected that any predictions made concerning the properties of an even-even alpha emitter in the region above lead should be quite accurate. On the other hand, predictions of the detail properties of the other mass types are not, in general, considered to be as reliable.

The systematics of the alpha emitters in this region are sufficiently well established to permit two general rules to be stated. These rules are considered to apply only to alpha emitting isotopes, either natural or artificially produced, which contain 128 or more neutrons.

1. In any element, the less the mass number, the shorter the alpha half-life.
2. In the region below uranium, for constant A - 2Z, the less the atomic number, the greater the alpha particle energy.

It has been found that the cross section for the production of the shorter activity is greater than that for the longer. The possibility that the two half-lives are associated with the same element is thus eliminated by rule 1. Also, from rule 2, the two activities are not members of the same decay series. It may be concluded then, that the two periods produced belong to an isotope of thorium and a new isotope of protactinium. The most probable mass assignments are to ${}_{90}Th^{224}$ and ${}_{91}Pa^{225}$. Of the short lived alpha activities that may be expected by the proton bombardment of thorium, these two require the least amount of energy of the incident protons and, therefore, may be expected to appear first as the energy of the bombarding particles is increased.

Predictions for the value of the half-life of Th^{224} have been made by two authors, Meinke(30,33) and Kaplan(10). The former has predicted the half-life as of the order of one second, and the latter as 0.6 second. The prediction drawn from Plate I agrees with that of Kaplan. Meinke has predicted the half-life of Pa^{225} as roughly 10 seconds, while the plot of Plate I indicates perhaps 2-3 seconds.

The conclusions to be drawn from these predictions are that the assignment of the 0.57 second activity should be made to Th^{224}, and the 2.0 second period to Pa^{225}. Other considerations must be taken into account before the assignment can be made finally.

2. The reactions involved in the production of these two isotopes are

$$Th^{232}(p,p8n)\ Th^{224}$$
$$Th^{232}(p,8n)\ Pa^{225}.$$

In addition to the half-lives, it has been shown that a value for the ratio of the cross sections for producing these isotopes could be obtained from the evaluation of the composite decay. Equation (13) gave,

$$\frac{\sigma(0.57 \text{ sec.})}{\sigma(2.0 \text{ sec.})} = 3.5 \pm 1 \quad .$$

The production of Th^{224} requires the emission of an additional particle to that required to produce Pa^{225}. The thresholds for these reactions may be calculated making use of the masses of the heavy isotopes as published by M. Stern(34). The mass difference between Th^{224} and Pa^{225} may be calculated from the difference between Pb^{208} and Bi^{209} by making use of the alpha disintegration energies of these chains. Although in the latter case these energies must be estimated, the estimates will not be in error by as much as 1 Mev.

The results of this calculation show that the threshold for production of Th^{224} is 47.1 Mev. and for Pa^{225}, 45 Mev. These are absolute thresholds and if the emitted particles are to have any kinetic energy, the actual observed threshold will be considerably higher.

In the case of charged particle emission from the compound nucleus, the effect of the coulomb barrier is important. It has been shown theoretically(35) that the evaporation model of nuclear reactions introduces a prohibition factor for the emission of a proton in competition with a neutron, of $e^{\frac{E_B^*}{T}}$ where E_B^* is 0.72 times the barrier height in Mev. and $\underline{T}$ is the nuclear temperature in Mev. The quantity $\underline{T}$ is given by $T = \sqrt{e_o E_p}$, where $\epsilon_o = \frac{1.40}{(A - 40)^{\frac{1}{2}}}$ and E_p is the excitation energy of the compound nucleus. In the present case, A = 233 and E_p = 90 Mev., and the prohibition factor is ~ 3. This aspect of the model has not been verified at high bombarding energies and it is only possible to conclude that the emission of a proton under these circumstances is not too improbable.

When the compound nucleus has been formed, as successive neutrons are evaporated, the binding energy per neutron will increase. At the end of eight neutron evaporations, it is quite probable that the excitation energy of the compound nucleus will not permit a further neutron to be emitted, but will be sufficient to allow a proton to escape with a few Mev. In competition with proton evaporation will be gamma ray emission. Provided the proton has sufficient energy to penetrate through the barrier in a short time, this mode of decay will be a large factor in the formation of the final nucleus.

The ratio of the cross sections is somewhat higher than might be expected from the above discussion, but the fact that at 90 Mev. there is more than enough energy to make the reaction take place in spite of the barrier, is sufficient reason to accept this value for the ratio as being close to the correct one.

That the evaporation model does not describe completely the process of nuclear reactions at high energies, has been pointed out by Serber[36]. In the case where the energy of the incident particle is of the order of 100 Mev. or greater, the compound nucleus may not be formed in the same manner that it is at lower excitation energies. At this energy the mean free path of the entering nucleon in the nucleus becomes important, and the probability of the transfer of a large fraction of the energy of the incident particle to one nucleon alone, the latter being ejected, is increased. The energy of the emitted nucleon would vary from a few Mev.,perhaps,up to the maximum available, and would leave the residual nucleus in a state of excitation similar to that assumed in the compound nucleus theory. This residual nucleus would then proceed to the final nucleus by evaporation.

The interaction of high energy neutrons with heavy nuclei has been investigated by Goldberger[37]. The energy distribution of particles emerging immediately after the bombardment of a heavy nucleus, shows a continuous distribution up to the maximum energy of the incident neutron. The distribution decreases regularly with increasing energy of emitted particle except near the maximum of the incident particle, where there is an increase. Since the interaction of protons of high energy with a heavy nucleus should not be too different, it is apparent from the results of Goldberger that the evaporation model does not suffice entirely to interpret nuclear reactions at high energies.

A further reaction which has been observed by J. Hadley and H. York[38] during the bombardment of nuclei with high energy neutrons, is the emission of fast deuterons. The energy of the ejected deuterons is, on the average, about one-half of the energy of the entering neutron. Total cross sections for the production of these deuterons from carbon, copper and lead are given. In the case of lead, which should correspond roughly to thorium, the cross section is 23 milli-barns. Owing to the fact that there are approximately two neutrons for every proton in a heavy nucleus, it might be expected that in the case of 90 Mev. proton bombardment, the cross section would be twice as great. The cross section for the emission of a fast proton was found to be 100 milli-barns for incident neutrons. On the same basis, the cross section for fast neutron emission under proton bombardment should be 200 milli-barns.

The experiments of Goldberger and Hadley and York verify that even in the heavy nuclei, the formation of the compound nucleus under 90 Mev. bombardment may not be considered as the only reaction process.

At the moment there is not sufficient experimental information available to estimate in what percentage of the reactions the compound nucleus is formed directly.

The qualitative arguments given above serve only to indicate that the (p,pxn) reaction could proceed equally as favourably as the (p,xn) in this region and under these circumstances. The fact that the cross section ratio favours the former is not too surprising.

3. The alpha particle energies of the Th^{224} series have been measured previously[33]. The half-lives of the members of this group have not been determined, but the existence of the chain is established from measurements on the parent nucleus U^{228}. The alpha particle decay energies have been given[32] as

$$Th^{224} \xrightarrow{7.13} Ra^{220} \xrightarrow{7.43} Em^{216} \xrightarrow{8.01} Po^{212} \xrightarrow{8.77} Pb^{208}$$

If this series is present as a contamination in the energy histogram, it is important to know the ratio of the number of Th^{224} disintegrations to the number of Pa^{225} disintegrations that have taken place while the source was next to the nuclear emulsion.

The last peak in the distribution has been assigned to At^{213} from systematics. From the number of particles in the peak immediately preceding At^{213}, which corresponds in energy to Po^{212}, it is possible to deduce that there are at least as many Po^{212} alpha particles present as At^{213}, with a good probability that the ratio should be two.

Now the ratio of the number of tracks on the plate is given by

$$\frac{N_1}{N_2} = \frac{S_1(1 - e^{-\lambda_1 T})(1 - e^{-\lambda_1 t})}{S_2(1 - e^{-\lambda_2 T})(1 - e^{-\lambda_2 t})} \quad \ldots\ldots\ldots\ldots\ldots\ldots(14)$$

where N_1 and N_2 are the number of Th^{224} and Pa^{225} tracks respectively, and the remaining symbols are as defined above. Since both the bombarding time and the recording time were three seconds,

$$\frac{N_1}{N_2} = \frac{S_1(1 - e^{-3\lambda_1})^2}{S_2(1 - e^{-3\lambda_2})^2}$$

The value of the ratio $\frac{S_1}{S_2}$ is obtained from equation (9), and insertion of the numerical values yields

$$\frac{N_1}{N_2} = 2.3$$

The ratio obtained in this manner is not inconsistent with that estimated from Figure 4.

The result of subtraction of the Th^{224} series from the total number of alpha particles is shown in Figure 6. The alpha particle energies of the members of this new series are determined from this corrected histogram.

The new series collateral to the neptunium 4n + 1 series is assigned as

$$_{91}Pa^{225} \xrightarrow{6.9} {}_{89}Ac^{221} \xrightarrow{7.6} {}_{87}Fr^{217} \xrightarrow{8.3} {}_{85}At^{213} \xrightarrow{9.2} {}_{83}Bi^{209}$$

Plate VIII shows the table of the isotopes in this region with the new additions included.

V. CONCLUSIONS

It has been found experimentally that the bombardment of thorium with 90 Mev. protons results in the production of two alpha active isotopes decaying with half-lives of 0.57 ± 0.15 sec. and 2.0 ± 0.3 sec. From the systematics of the alpha emitters, the consistency of which has been established in this region, it has been possible to assign these two periods to ${}_{90}Th^{224}$ and ${}_{91}Pa^{225}$ respectively. The presence of the thorium isotope has been established previously[30], but no half-life determinations have been reported.

The protactinium isotope is a member of a new chain of alpha emitters, collateral to the artificially produced neptunium 4n + 1 series. The energy of the alpha particle emitted by this isotope, together with the energies of the alphas from its three alpha emitting daughters, have been measured. The decay chain is assigned as follows.

$$ {}_{91}Pa^{225} \xrightarrow{6.9} {}_{89}Ac^{221} \xrightarrow{7.6} {}_{87}Fr^{217} \xrightarrow{8.3} {}_{85}At^{213} \xrightarrow{9.2} {}_{83}Bi^{209} $$

The third daughter of Pa^{225} is astatine mass number 213. Interest is centered in the energy of the alpha particle emitted from this isotope owing to the fact that it contains 128 neutrons, which are two more than a closed shell. It had been estimated[8] that the alpha disintegration energy of this nucleus would be 9.2 Mev. The energy obtained from this experiment is 9.3 Mev., which is in good agreement with the predicted value.

VI. ACKNOWLEDGEMENTS

The author wishes to express his gratitude and appreciation to Dr. J. S. Foster for his continued interest and invaluable suggestions during the course of this research.

It is also a pleasure to extend thanks to Drs. S. W. Breckon and W. M. Martin and Mr. R. H. Mills for their co-operation in making available, and aiding in the operation of, the rapid target extractor, which has made this experiment possible. In addition, the discussions held with Dr. Breckon have aided greatly in the interpretation of the experimental results obtained with the extractor.

The members of the Radiation Laboratory in general have been very helpful, and in particular Mr. S. Doig has been most co-operative in his capacity of foreman of the machine shop.

This work was performed with the aid of a grant from the National Research Council of Canada.

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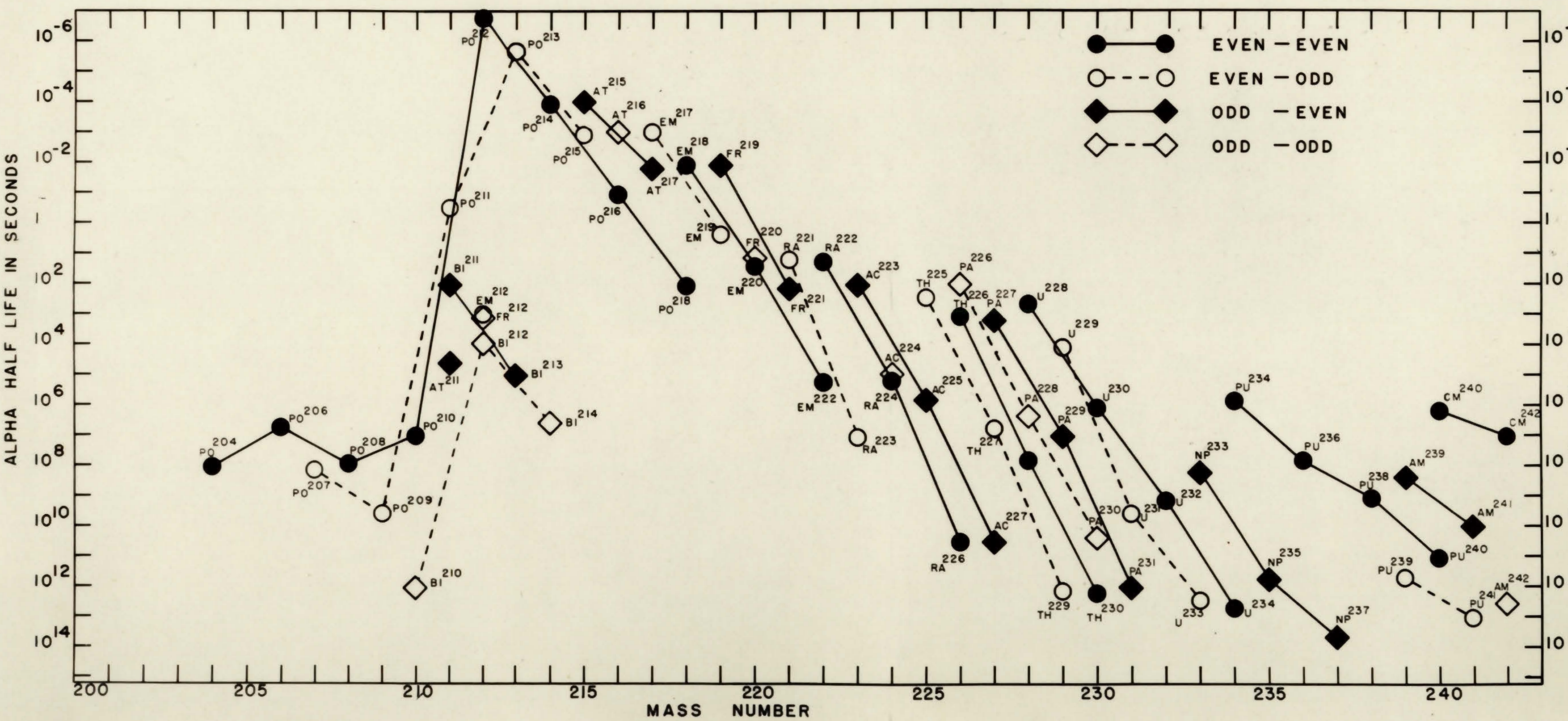
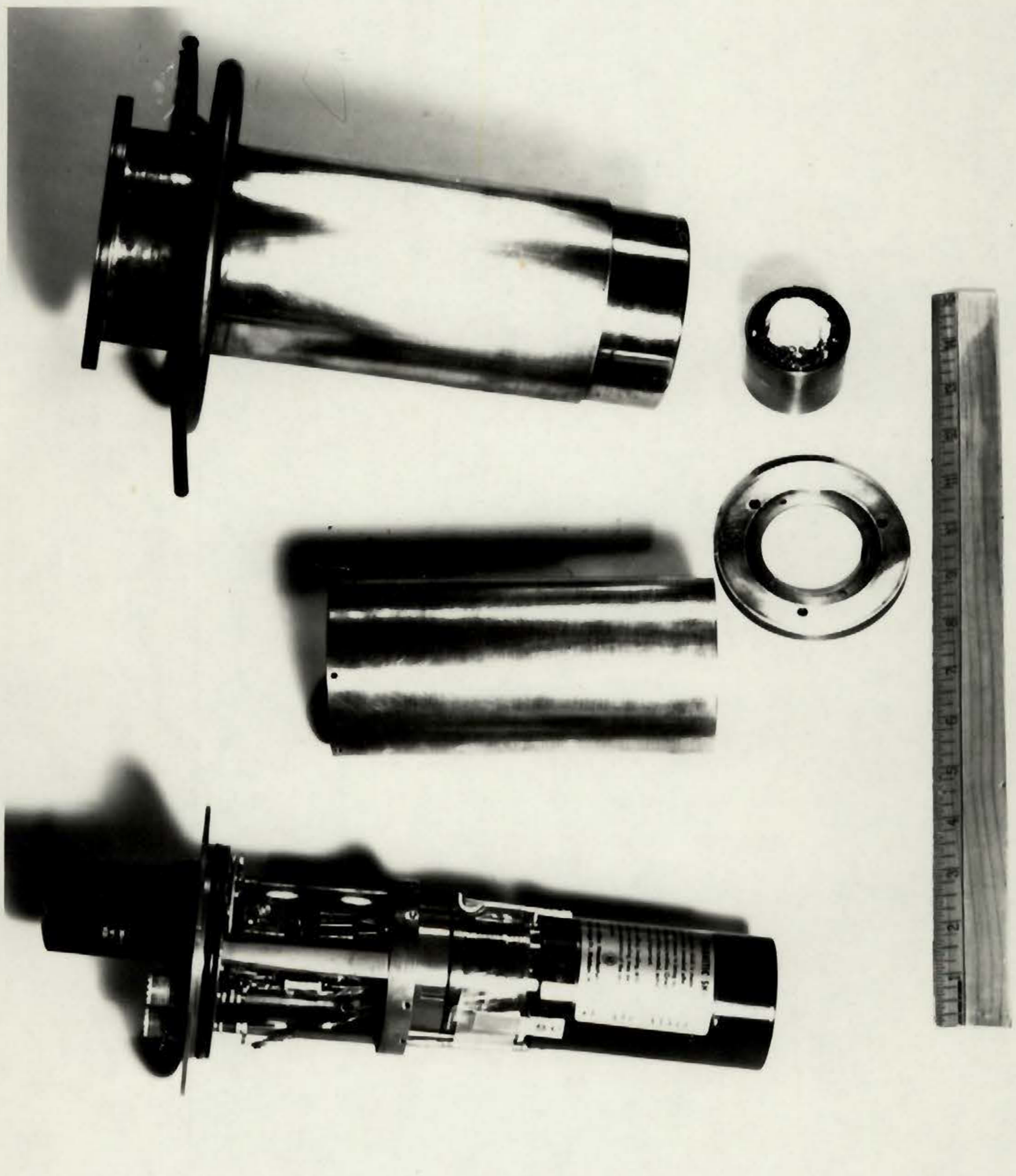


EVEN — EVEN
EVEN — ODD
ODD — EVEN
ODD — ODD
ALPHA HALF LIFE IN SECONDS
MASS NUMBER

PLATE I



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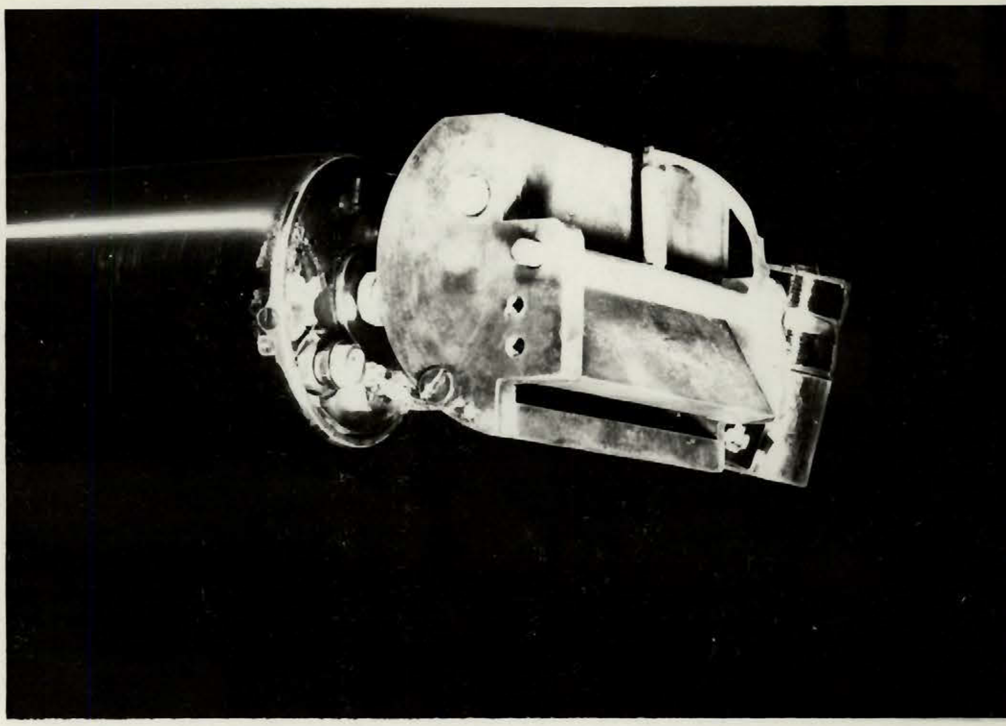

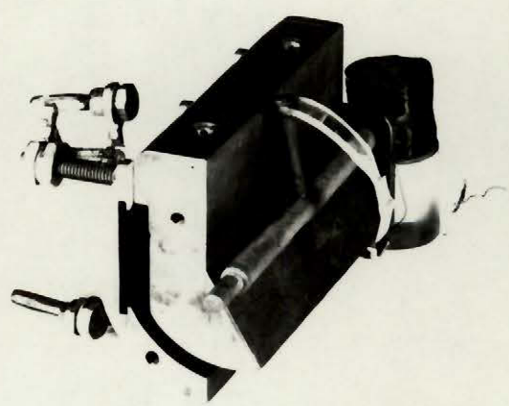

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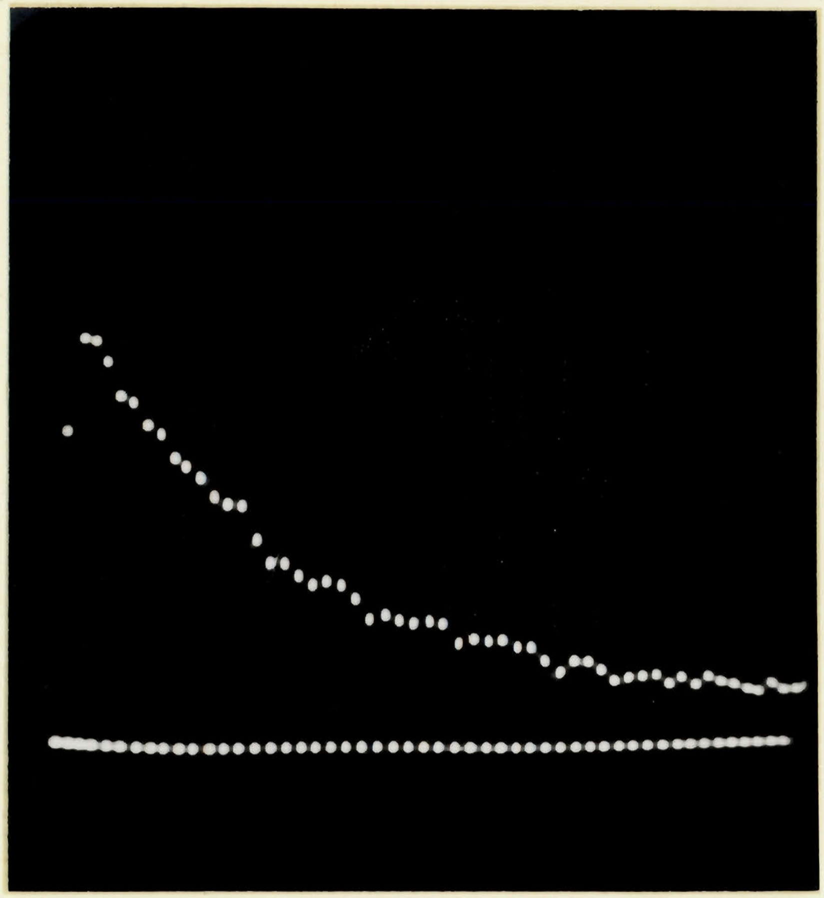


PLATE IV

Th-3-5. Time base speed 5.4 secs.
10.0_8 marker pips per second.
Half-life 1.0_7 secs.

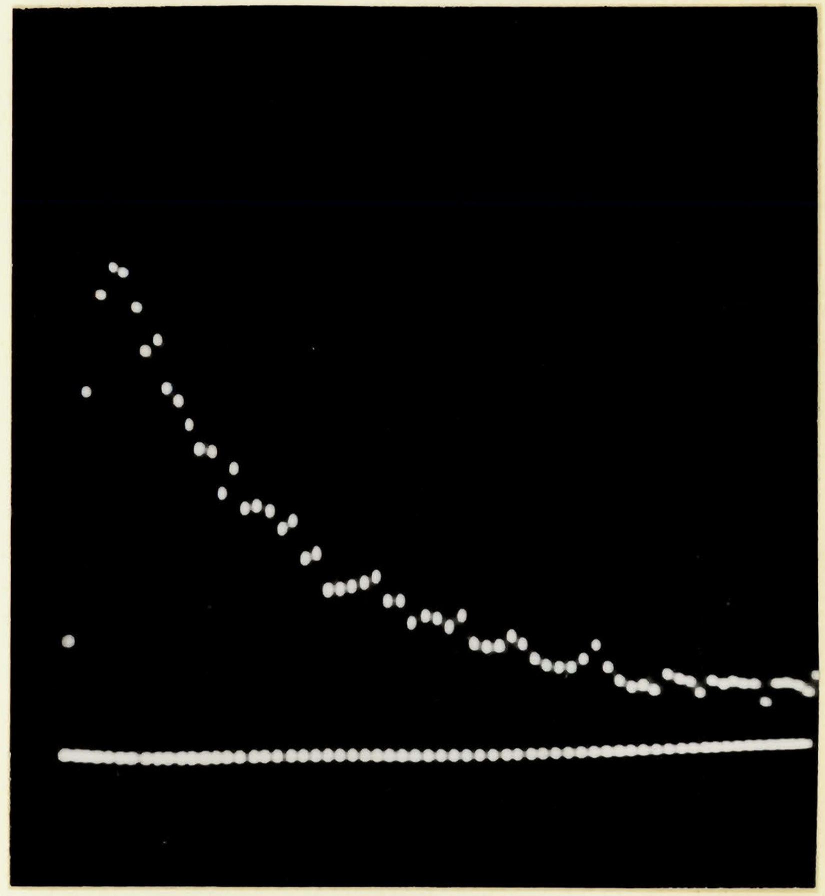


PLATE V

Th-4-4. Time base speed 6 secs.
10.0_1 marker pips per second.
Half-life 1.1_3 secs.

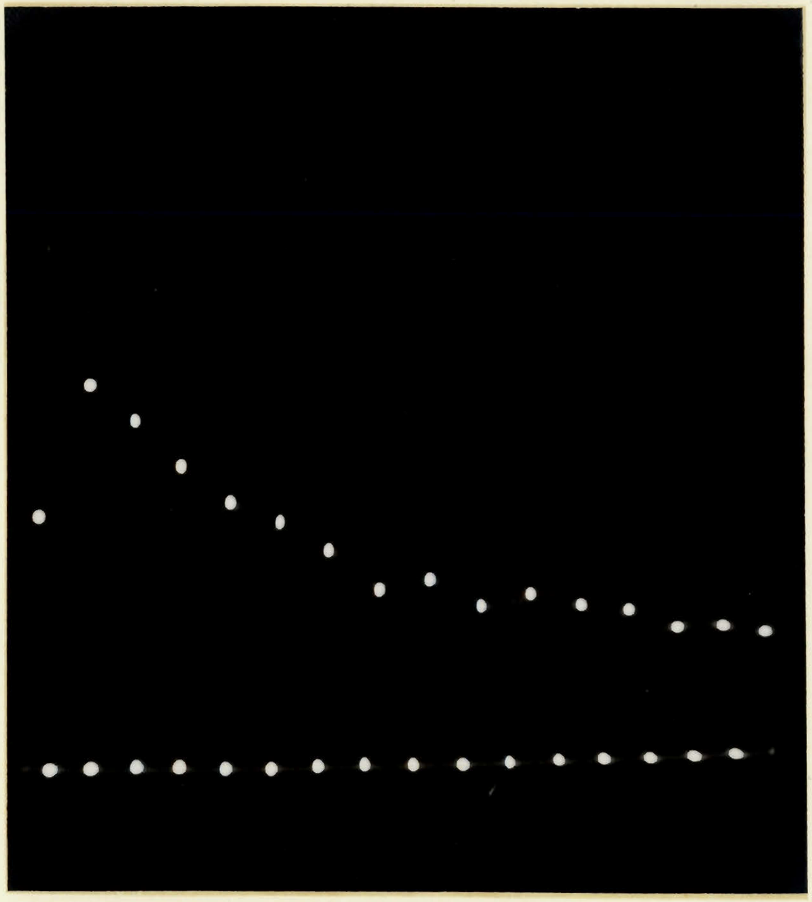


PLATE VI

Th-4-37. Time base speed 8 secs.
2 marker pips per second.
Half-life 2.2_0 secs.

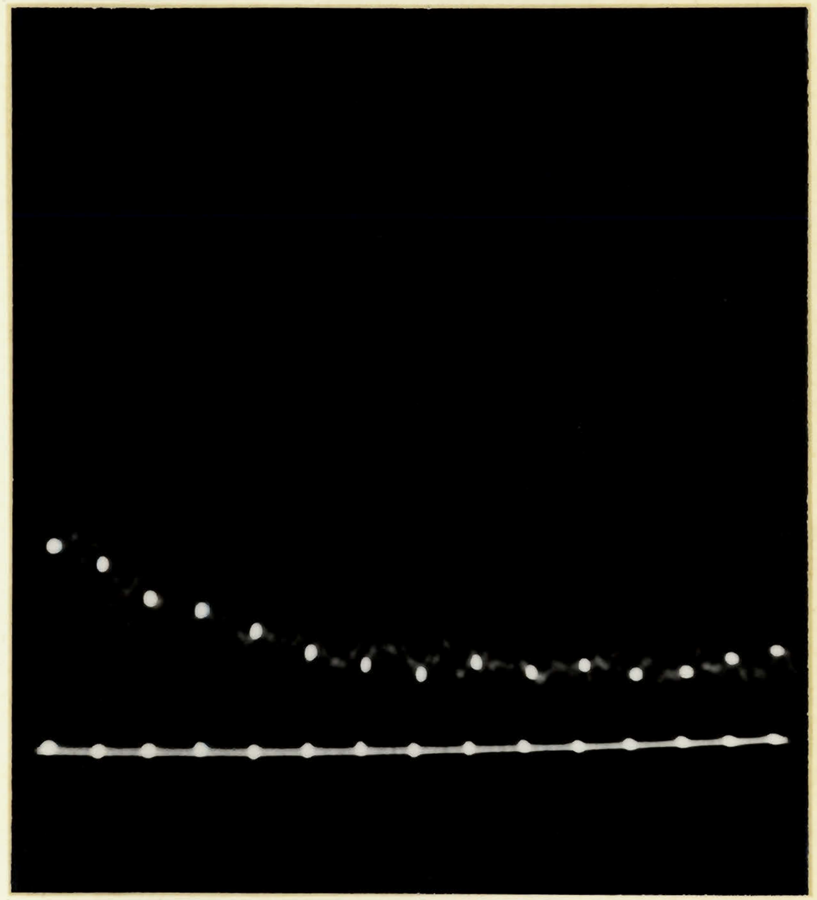


PLATE VII

Th-5-12. Time base speed 8.2 secs.
1.8_3 marker pips per second.
Half-life 1.3_6 secs.

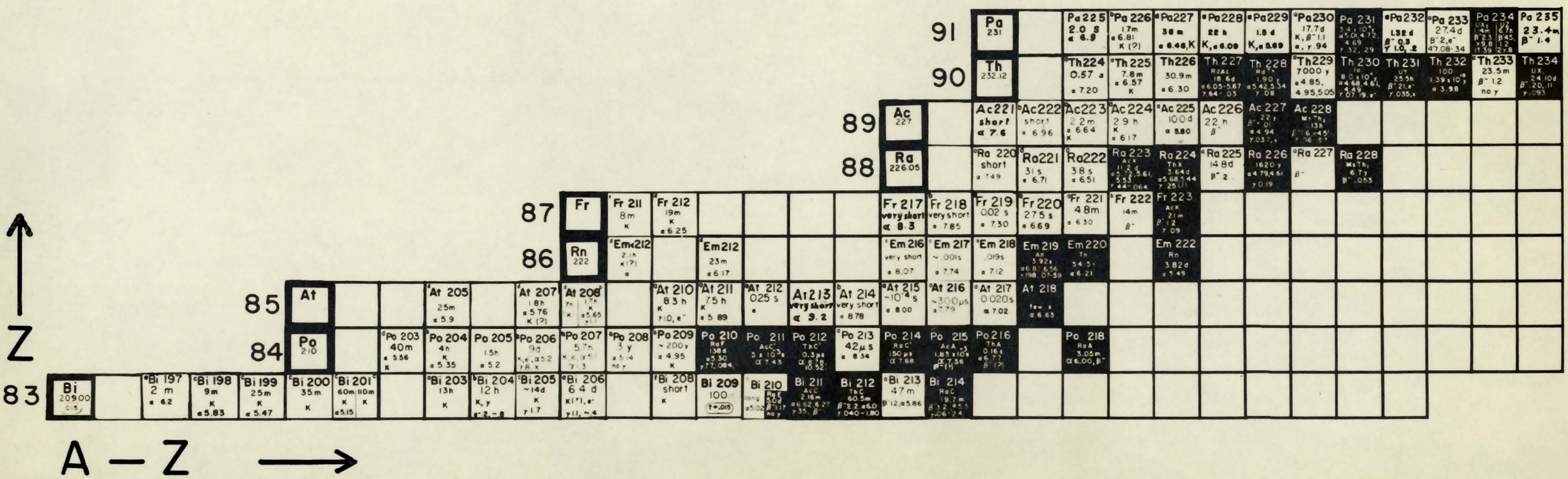

91
90
89
88
87
86
85
84
83
Pa
231
Th
232.12
Ac
227
Ra
226.05
Fr
Rn
222
At
Po
210
Bi
209.00
Z
A – Z

PLATE VIII

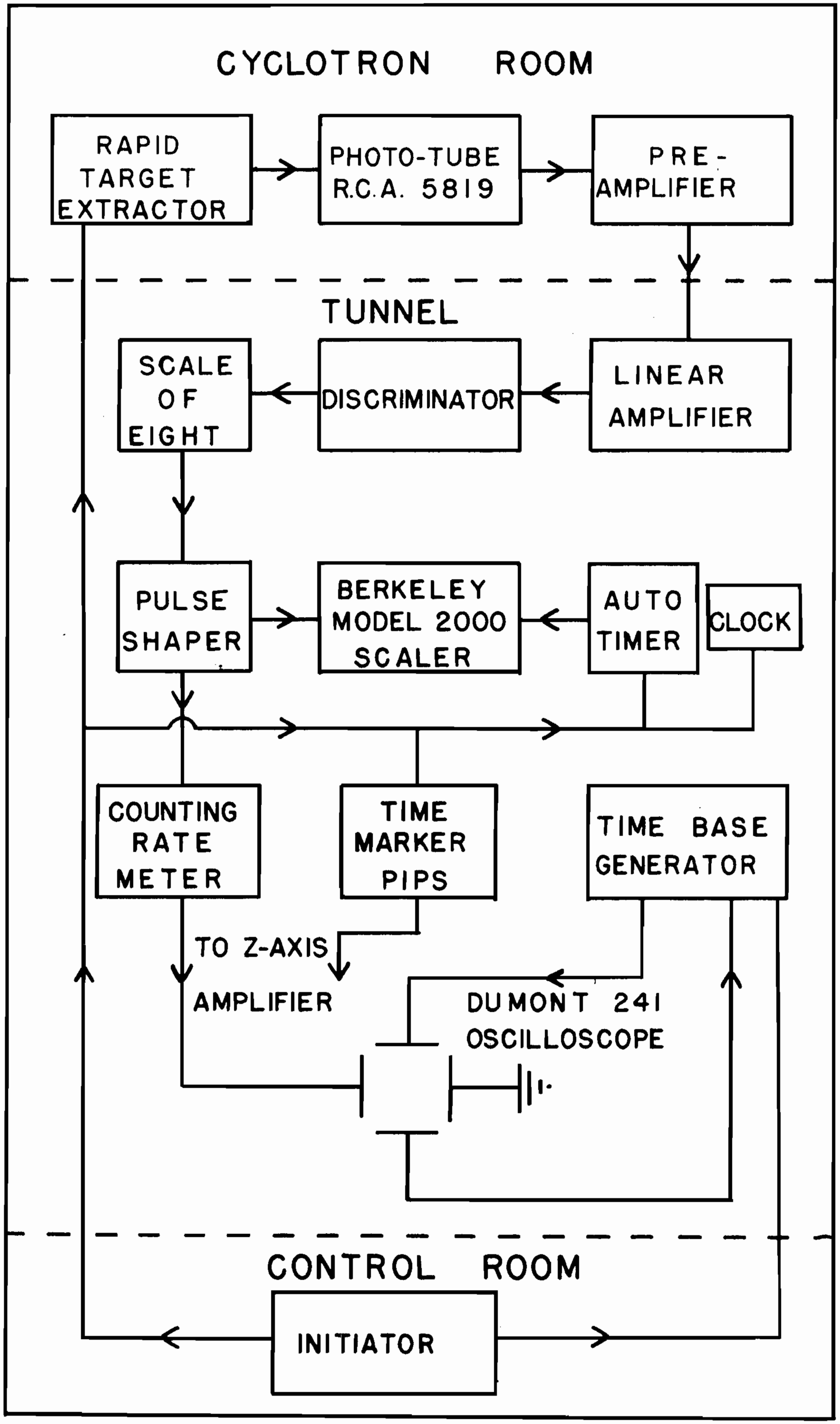
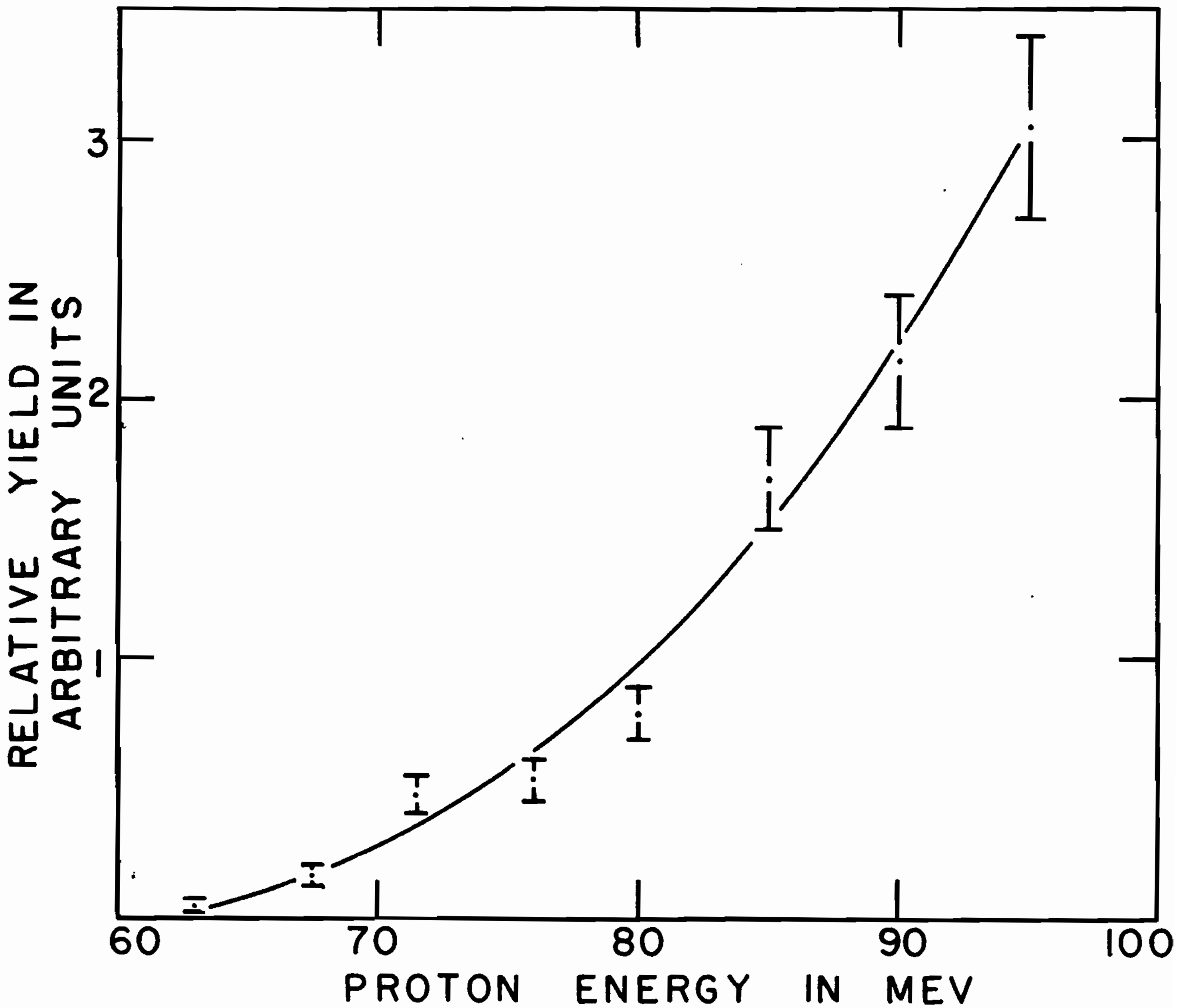
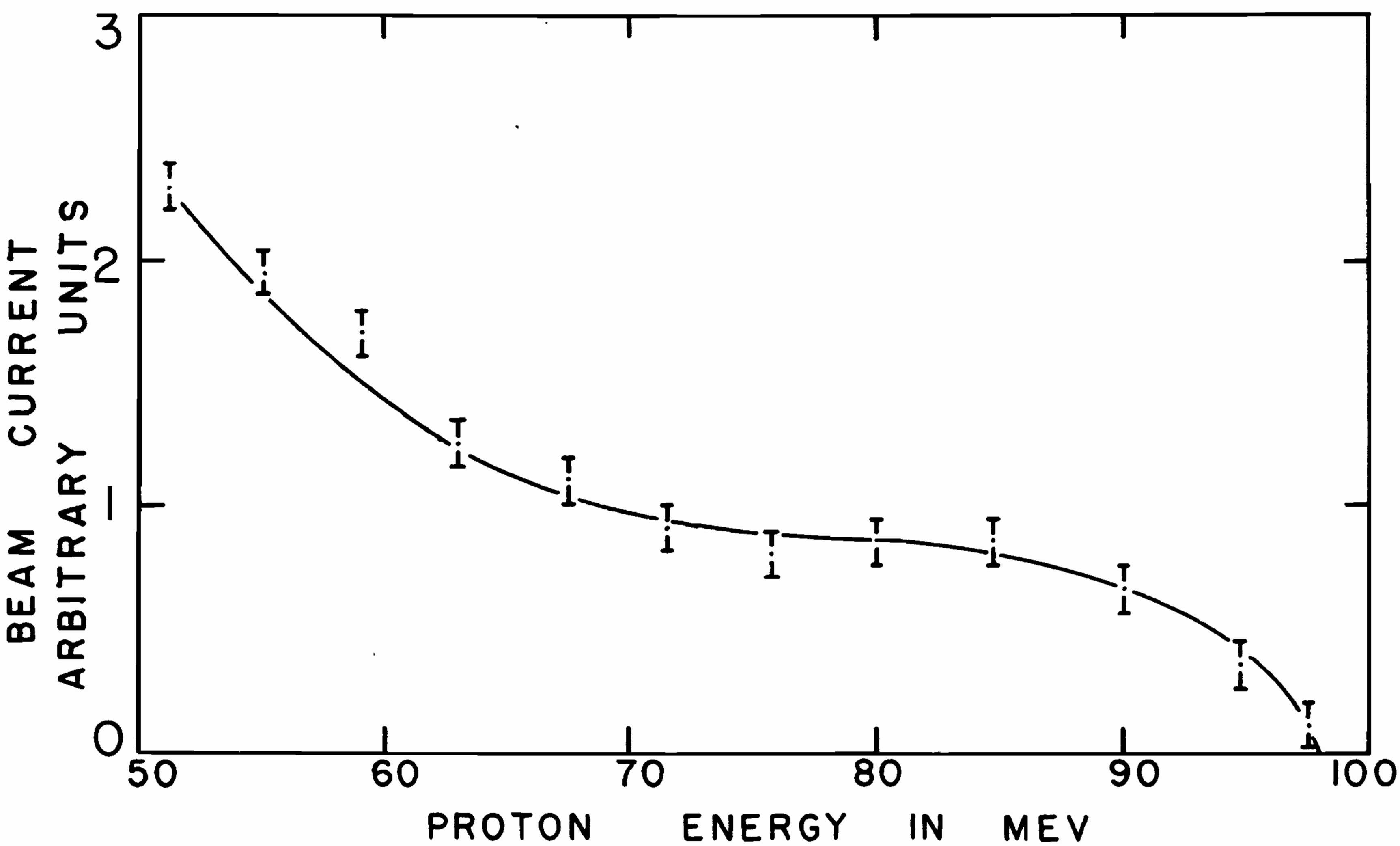

CYCLOTRON ROOM
RAPID TARGET EXTRACTOR
PHOTO-TUBE R.C.A. 5819
PRE-AMPLIFIER
TUNNEL
SCALE OF EIGHT
DISCRIMINATOR
LINEAR AMPLIFIER
PULSE SHAPER
BERKELEY MODEL 2000 SCALER
AUTO TIMER
CLOCK
COUNTING RATE METER
TIME MARKER PIPS
TIME BASE GENERATOR
TO Z-AXIS AMPLIFIER
DUMONT 241 OSCILLOSCOPE
CONTROL ROOM
INITIATOR

FIGURE I



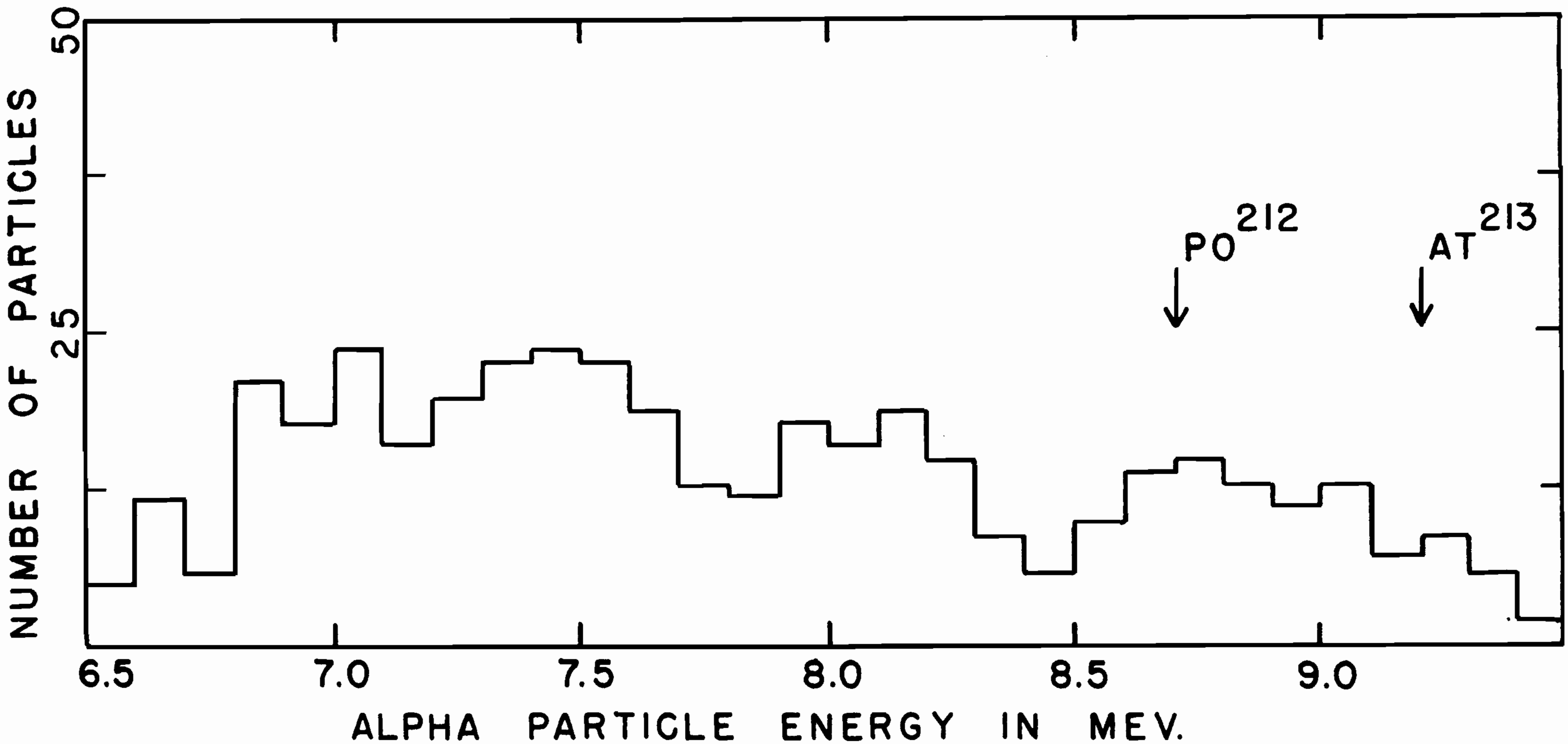
RELATIVE YIELD IN ARBITRARY UNITS
3
2
1
60
70
80
90
100
PROTON ENERGY IN MEV

FIGURE 2



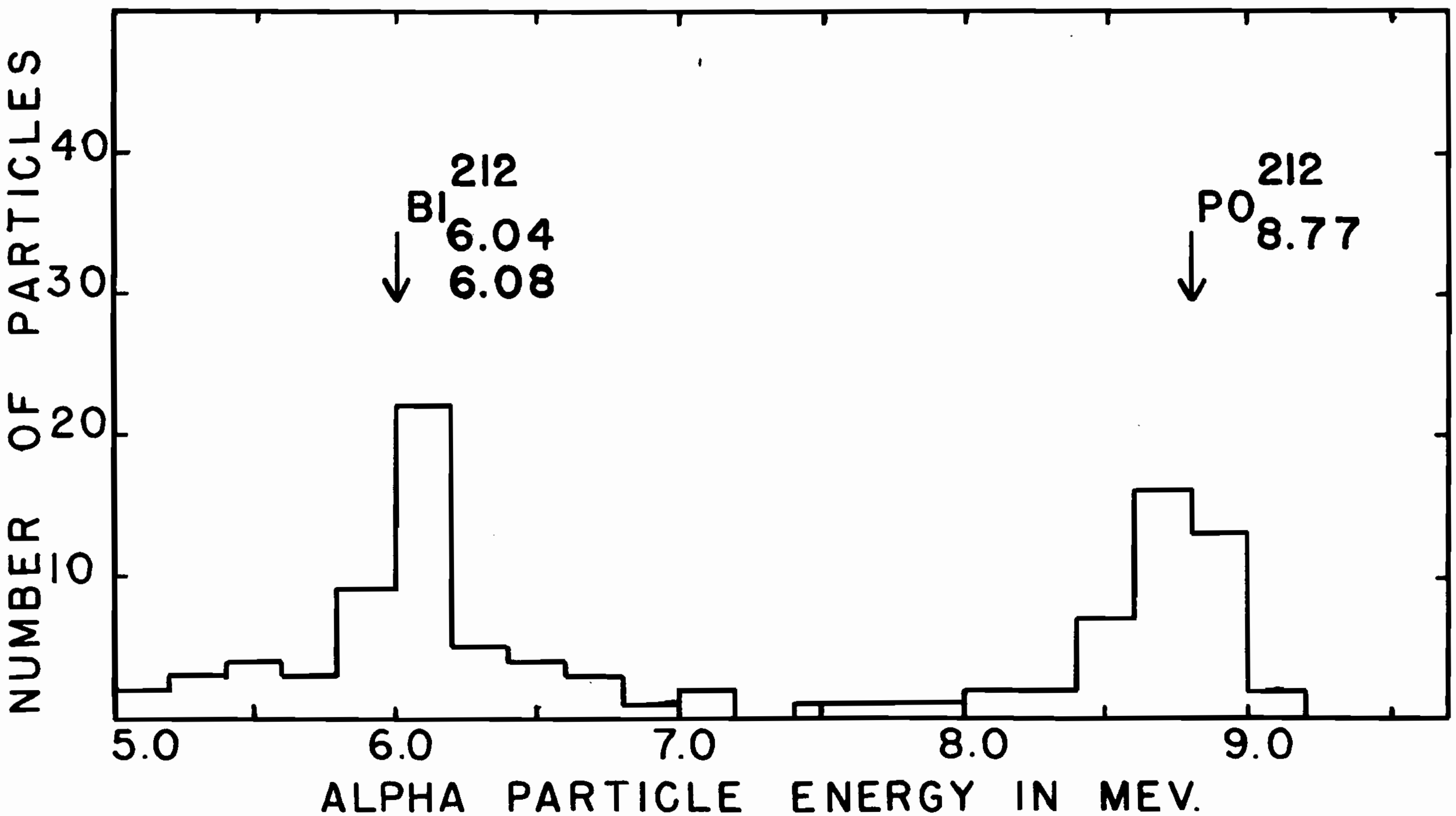
3
2
1
0
BEAM CURRENT
ARBITRARY UNITS
50
60
70
80
90
100
PROTON ENERGY IN MEV

FIGURE 3



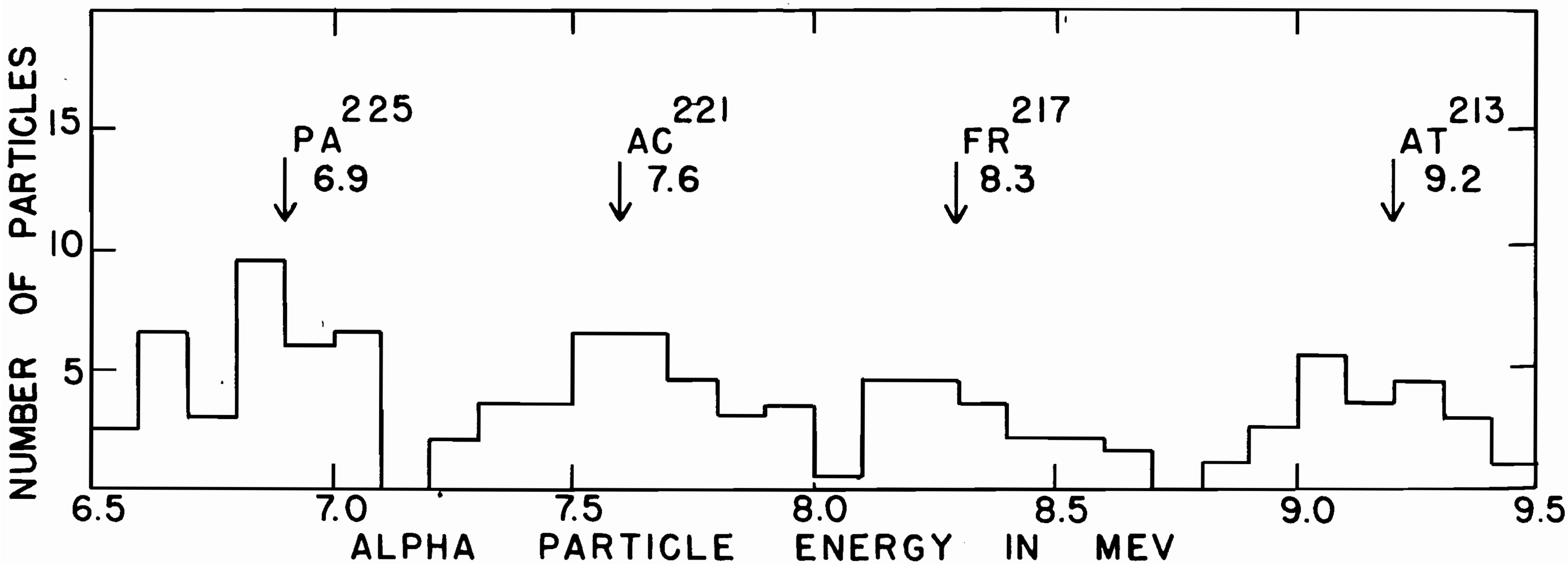
50
25
NUMBER OF PARTICLES
PO^{212}
AT^{213}
6.5
7.0
7.5
8.0
8.5
9.0
ALPHA PARTICLE ENERGY IN MEV.

FIGURE 4



BI 212
6.04
6.08
PO 212
8.77
NUMBER OF PARTICLES
10
20
30
40
5.0
6.0
7.0
8.0
9.0
ALPHA PARTICLE ENERGY IN MEV.

FIGURE 5



NUMBER OF PARTICLES
15
10
5
PA 225
6.9
AC 221
7.6
FR 217
8.3
AT 213
9.2
6.5
7.0
7.5
8.0
8.5
9.0
9.5
ALPHA PARTICLE ENERGY IN MEV

FIGURE 6