THE ALPHA BRANCHING OF ACTINIUM-227 AND THE HALF LIFE OF FRANCIUM-223

By

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CHAPTER I

INTRODUCTION

Actinium Series

Actinium-227 and Francium-223 belong to the (4n + 3) natural radioactive series. Ever since the early days of radioactivity, the heavy nuclides have been known to be radioactive, and so the actinium series, having mass numbers from 207 to 235, has as its main mode of decay, alpha particle emission. The nature of the actinium series was a source of much controversy during the first third of this century (1,2,3). The independence of this series from the uranium (4n + 2) series had not been established and the mass numbers of the parents of actinium, viz., protactinium, thorium, and uranium had not been assigned. With the invention of the mass spectrograph by F. W. Aston, the end product of the series was established as Pb-207(4). This fact was also determined chemically in 1932 by A. v. Grosse (5). This then fixed the atomic weight of protactinium as being Pa-231 since six alpha emissions occur between these two nuclides (207 + 6 x 4). However Aston's measurements on a uranium source failed to show any isotope of uranium other than U-238. It was not until 1935 when A. J. Dempster developed a new type of mass spectrograph that the isotope of uranium of atomic weight 235 was found and established to be the parent of the actinium series (6).

Figure 1 shows the decay scheme for the actinium series and gives

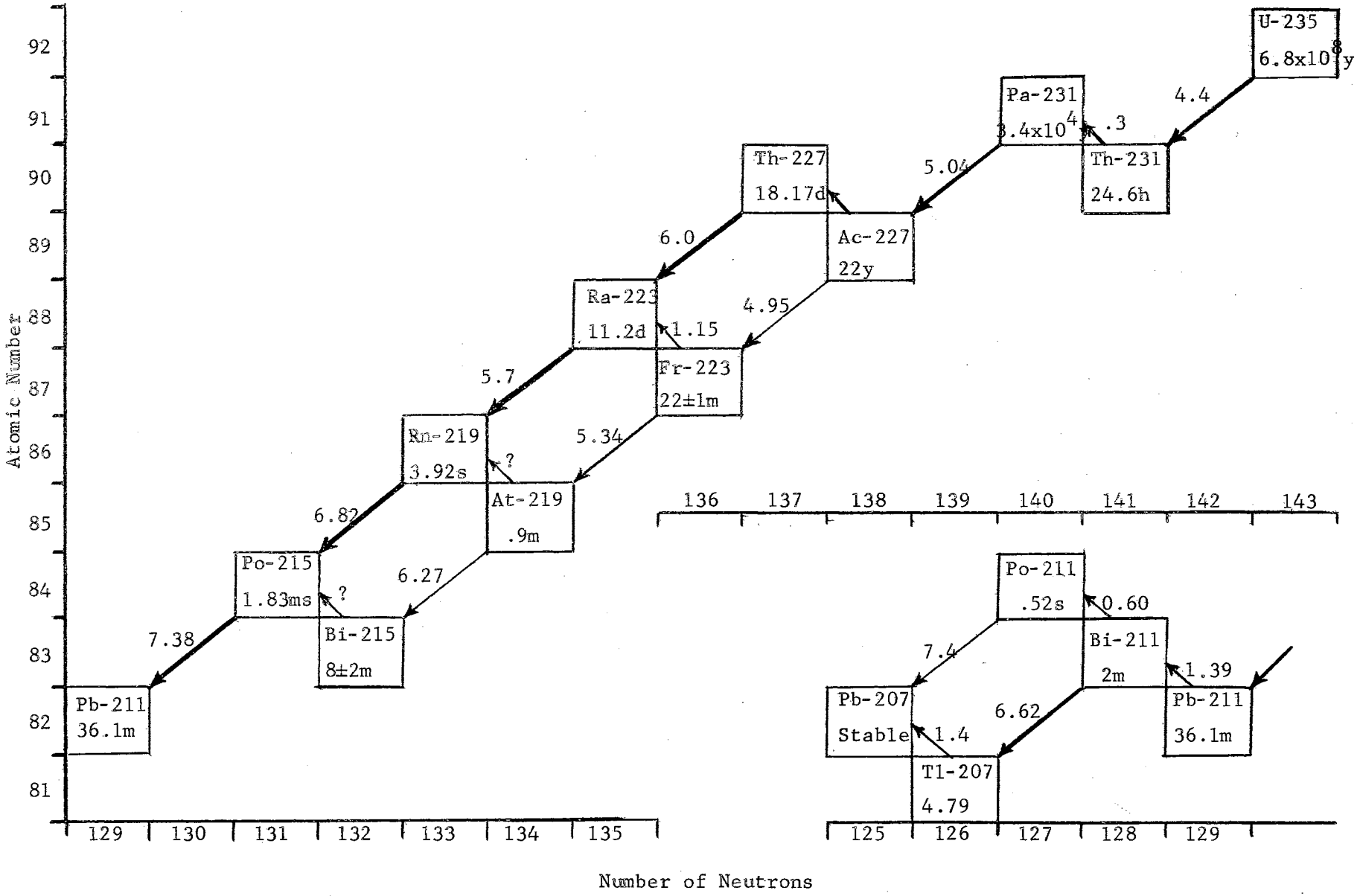

Figure 1. The Actinium Series

the various half lives and particle energies of its members. The heavy lines indicate the most probable mode of decay while the lighter lines indicate the alternate decay schemes. U-235 has a half life of 6.8×10^8 years (7). This nuclide decays with the emission of a 4.4 mev alpha particle (8) to Th-231. Th-231 in turn has a much shorter half life of 24.6 hours (9) and decays by beta emission to Pa-231. This alpha active nuclide has a half life of 3.43×10^4 years (10) and decays into the nuclide of interest here, Ac-227.

Actinium-227 History

Ac-227 has been the subject of much investigation. While Figure 1 shows an alpha energy of 4.95 mev for actinium, there is really a fine structure in the alpha emission (11). The energies in this fine structure are, however, close together as 48% of the decays take place by the emission of a 4.949 mev alpha particle and 36% of the decays take place by the emission of a 4.936 mev alpha particle as can be seen below in Figure 2.

Energy in mev	%
1. 4.949	48.7
2. 4.936	36.1
3. 4.866	6.9
4. 4.849	5.5
5. 4.786	1.0
6. 4.795	1.8
7. 4.728	0.1
8. 4.704	0.4
9. 4.517	0.2

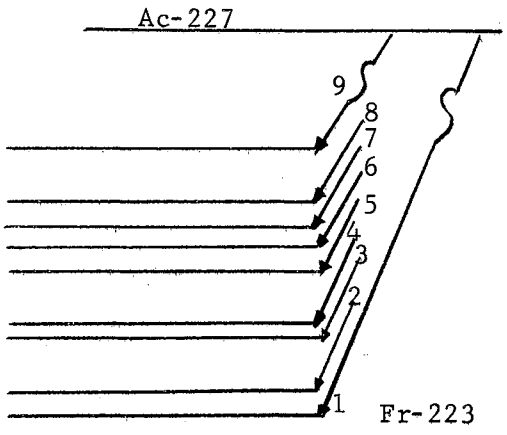

Figure 2. Fine Structure in the Alpha Particle Emission of Ac-227

The beta and gamma energies are also well known (11) and the half life of actinium has been found to be 22 years (12,13). The only real gap in the picture of actinium lies in its branching to Th-227 and Fr-223 as shown in Figure 1. This branching was first noticed by Mayer, Hess and Paneth (14) in 1914, but its actual existence was still in question until 1939 when Percy isolated the alpha daughter, Fr-223 (15). The value of the branching could only be roughly estimated since the measurement depended upon the beta activity of the sample and no direct measure of the alpha particles were made. Up to the present time only one actual result of a branching measurement has been reported, and that was the 1.25% branching obtained by Peterson and Ghiorso (16). The main endeavor of this research was to make another measurement of this branching ratio and thereby also calculate the partial alpha half life associated with Ac-227.

Properties of Francium

In 1955, J. P. Adloff reported a series of measurements on francium properties (17) and later in 1956, M. Perey and J. P. Adloff repeated these attempts (18). The value obtained for the half life was 22±1 minute and the alpha/beta branching was given a value of approximately 6×10^{-5}. Due to the small branching ratio nearly all the francium atoms decay to Ra-223 by beta emission of end point energy 1.15 mev (19). The other francium atoms decay by a 5.34 mev alpha particle to At-219. This nuclide has a short half life of .9 minutes and decays nearly 100% to Bi-215 (20) which then rejoins the main decay scheme by beta emission.

To prepare a Fr-223 source suitable for a half life measurement,

the francium must be separated from all other series members. It was felt that if some Pb-211 contamination was present in the sources previously used to determine this half life, the recorded result would be too large since the Pb-211 has a 36 minute half life. The alpha spectrometry equipment that was available provided an excellent method for making sure this contaminate was not present and therefore a check on the half life of Fr-223 was made.

THEORY

Actinium Branching

The alpha branching of an element can be found from the equation

$$\text{Branching ratio} = \frac{\lambda_\alpha}{\lambda_T} \tag{1}$$

where λ_α is the partial decay constant for alpha decay and λ_T is the total decay constant. If there are two modes of decay, alpha and beta emission, then $\lambda_T = \lambda_\alpha + \lambda_\beta$. Equation (1) can then be rewritten as

$$\text{Branching ratio} = \frac{\lambda_\alpha}{\lambda_\alpha + \lambda_\beta} = \frac{\lambda_\alpha/\lambda_\beta}{1 + \lambda_\alpha/\lambda_\beta} \tag{2}$$

so that if the ratio of the decay constants for an element's alpha and beta emission can be found, the branching ratio can be determined. These values were measured in this work using an alpha spectrometry technique.

In its natural state the actinium series is in secular equilibrium. The conditions for this type of equilibrium are that the parent nuclide be much longer lived than any of its daughters and that the material has been left alone for a time much longer than the half life. The half life of Ac-227 is 22 years while the longest lived daughter is

Th-227 which has an 18.17 day half life (21). Therefore the first condition given above is well satisfied.

The specific activity of an element is the number of disintegrations per gram per second taking place. The activity (disintegrations per second) of a radioactive nuclide is known to follow the exponential law

$$I_x(t) = I_x^o e^{-\lambda t} \tag{3}$$

where I_x^o is the initial activity and λ is a constant for that particular element and is called the decay constant. The observed activity of a radioactive element is proportional to the number of atoms (N) in the material; the relation between the two is

$$A = c\lambda N \tag{4}$$

where c is a proportionality factor depending upon the nature and efficiency of the detection instrument (22).

In a radioactive series such as actinium, the activity of the first member can be rewritten by combining Equations (3) and (4).

$$N_1\lambda_1 = N_1^o\lambda_1 e^{-\lambda_1 t} \tag{5}$$

N_1^o is the initial number of atoms of the first member and λ_1 is the decay constant of the first member. The activity of the second member of the series is given by

$$N_2\lambda_2 = N_1^o\lambda_{1(\beta)} \frac{\lambda_2}{\lambda_2 - \lambda_{1(\beta)}} \left[e^{-\lambda_1(\beta)t} - e^{-\lambda_2 t} \right] \tag{6}$$

if at t = 0 only the first member of the series exists (22). The decay constant for the second member of the series, thorium, is λ_2 and the decay constant for the first member has been written as $\lambda_{1(\beta)}$ since the equation as written above included only atoms decaying from actinium to thorium. This decay is by beta emission. The exponent of equation (6)

can be expanded in powers of λt giving

$$N_2\lambda_2 = N_1^o\lambda_{1\beta}\left(\frac{\lambda_2}{\lambda_2 - \lambda_{1\beta}}\right)\left[(\lambda_2 - \lambda_{1\beta})t + \tfrac{1}{2}(\lambda_1^2 - \lambda_2^2)t^2\right] \quad (8)$$

$$N_2\lambda_2 = N_1^o\lambda_{1\beta}\lambda_2\left[t - (\lambda_2 + \lambda_{1\beta})t^2/2\right] = N_1^o\lambda_{1\beta}\lambda_2 t' \quad (9)$$

where t' is the "reduced time" = $t - (\lambda_2 + \lambda_1)t^2/2$.

To apply these equations to the work here let:

A_1 be the number of observed actinium alpha particles per second;

A_2 be the number of observed actinium beta particles per second;

C_1 be the counting efficiency for actinium alpha particles per second;

C_2 be the counting efficiency for actinium beta particles per second.

Then, from Equation (4):

$$A_1 = C_1\lambda_{1\alpha}N_1 \quad (10)$$

$$A_2 = C_2\lambda_{1\beta}N_1 \quad (11)$$

and

$$\frac{A_1}{A_2} = \frac{C_1}{C_2}\frac{\lambda_{1\alpha}}{\lambda_{1\beta}} \quad (12)$$

Therefore if one can find A_1, A_2, and the counting efficiencies, $\lambda_{1\alpha}/\lambda_{1\beta}$ can be calculated and the branching ratio found from Equation (2). However c_2, the counting efficiency for beta particles, cannot be obtained accurately and so these two equations alone aren't enough. The beta decay product of actinium is Th-227 which as mentioned before is an alpha emitter. Letting A_3 equal the number of thorium alpha particles observed per second, another equation can be obtained

$$A_3 = c_3\lambda_2 N_2 \quad (13)$$

where c_3 is the counting efficiency for the alpha particles. The value of $\lambda_2 N_2$ from Equation (9) can be substituted into Equation (13) giving

$$A_3 = c_3 N_1^o \lambda_{1\beta} \lambda_2 t' \tag{14}$$

and

$$\frac{dA_3}{dt'} = c_3 N_1^o \lambda_{1\beta} \lambda_2 \tag{15}$$

Solving for $\lambda_{1\alpha}$ in Equation (10) and $\lambda_{1\beta}$ in Equation (15) gives the following two equations:

$$\lambda_{1\alpha} = \frac{A_1}{c_1 N_1} \tag{16}$$

$$\lambda_{1\beta} = \frac{dA_3/dt'}{c_3 N_1^o \lambda_2} \tag{17}$$

In Equation (5) the value of λ_1 is $9.838\ (10)^{-10}$ sec^{-1} and therefore for $t < 24$ hours

$$\lambda_1 N_1 \approx \lambda_1 N_1^o \tag{18}$$

or

$$N_1 \approx N_1^o \tag{19}$$

Dividing Equation (17) into Equation (16) and substituting in N_1 for N_1^o in (17) the following ratio is obtained.

$$\frac{\lambda_{1\alpha}}{\lambda_{1\beta}} = \frac{\dfrac{A_1}{c_1 N_1}}{\dfrac{dA_3/dt'}{c_3 N_1 \lambda_2}} \tag{20}$$

The counting efficiencies for the actinium alpha particles and the thorium alpha particles are very nearly equal and so their ratio very nearly one. Therefore the ratio of the partial decay constants is

$$\frac{\lambda_{1\alpha}}{\lambda_{1\beta}} = \frac{\lambda_2 A_1}{dA_3/dt'} \tag{21}$$

If the components of the right-hand side of this equation can be found then the branching ratio can be obtained from Equation (2).

A_1 is the number of observed actinium alpha particles per second. Since actinium has a half life of 22 years, the number of atoms decaying in a ten-minute timing period will be the same for any ten-minute run over the 24-48 hour counting time that a source was used. This was shown in Equations (18) and (19). The condition placed on Equation (6) was that at $t = 0$ only actinium atoms be present in the source. Therefore a freshly prepared source -- one which has had the daughters of actinium removed -- was made for each measurement of the branching. The data in this was recorded by a pulse height analyzer and since only actinium atoms were present at the beginning of a run, the value of A_1 could be obtained quite readily.

The other quantity in Equation (21) that had to be measured was dA_3/dt', the growth rate of thorium. As shown in Figure 1, thorium is the beta daughter of actinium and since each freshly prepared source contained only actinium at the beginning, the thorium began to immediately grow in for $t > 0$. This rate of growth is governed by Equation (9). If thorium itself were not radioactive it would grow in as a linear function of time. However since it too is radioactive, it begins to decay into Ra-223. So the rate of growth of thorium will be a linear function of the reduced time, t', instead of t alone, as given in Equation (9). This value for the rate of growth of thorium can be obtained by plotting the observed thorium counts against the reduced time, the slope of the straight line being the rate of growth and the needed value in Equation (21). One big advantage in taking the measurement of the branching ratio in this manner is that the two measurements

that are needed, namely the actinium alpha activity and the thorium growth rate, are performed simultaneously using the same source and the same geometry.

Francium Half Life

The half life of francium was found in the standard way using a Geiger-Mueller counter. A source containing only francium atoms is approximately 100% beta active ($\alpha/\beta = 6 \times 10^{-5}$) and so suitable counting was possible with the G-M counter. The log of the counts per second was plotted as a function of time and the half life obtained from the slope of the curve.

CHAPTER II

APPARATUS, SOURCES, AND EXPERIMENTAL PROCEDURE

Equipment and Circuits

The apparatus used to detect alpha particles in this work was a parallel plate grided ionization chamber similar to the one described by Karras and Nurmia (23,24). The chamber was 14 cm high and had an inside diameter of 12 cm. The inner parts consisted of a source plate, grid, and collector. The parts were made from brass and were nickel plated to aid in cleaning. As seen in Figure 3, the assembly was built up from the base of the chamber by the use of three metal supporting rods running the length of the chamber. Around these rods were some glass tubing which served as insulation. Slightly larger pieces of glass tubing (1 cm in outside diameter) were then placed around the other tubes, these serving as spacers and support for the grid and source plate. The collector was supported from the base by a 5.2 cm long lucite rod. The grid was separated 1.28 cm from the collector and the source plate was 1.96 cm from the grid. The source plate and grid were 11.17 cm in outside diameter while the collector was 7.5 cm in diameter. The grid was wound with 0.0254 mm wire, the wire spacing being one mm apart. The potential applied to the components was done by a divider circuit as described by Sievers (25). The collector was at a potential of 1200 volts while the grid's potential was 600 volts.

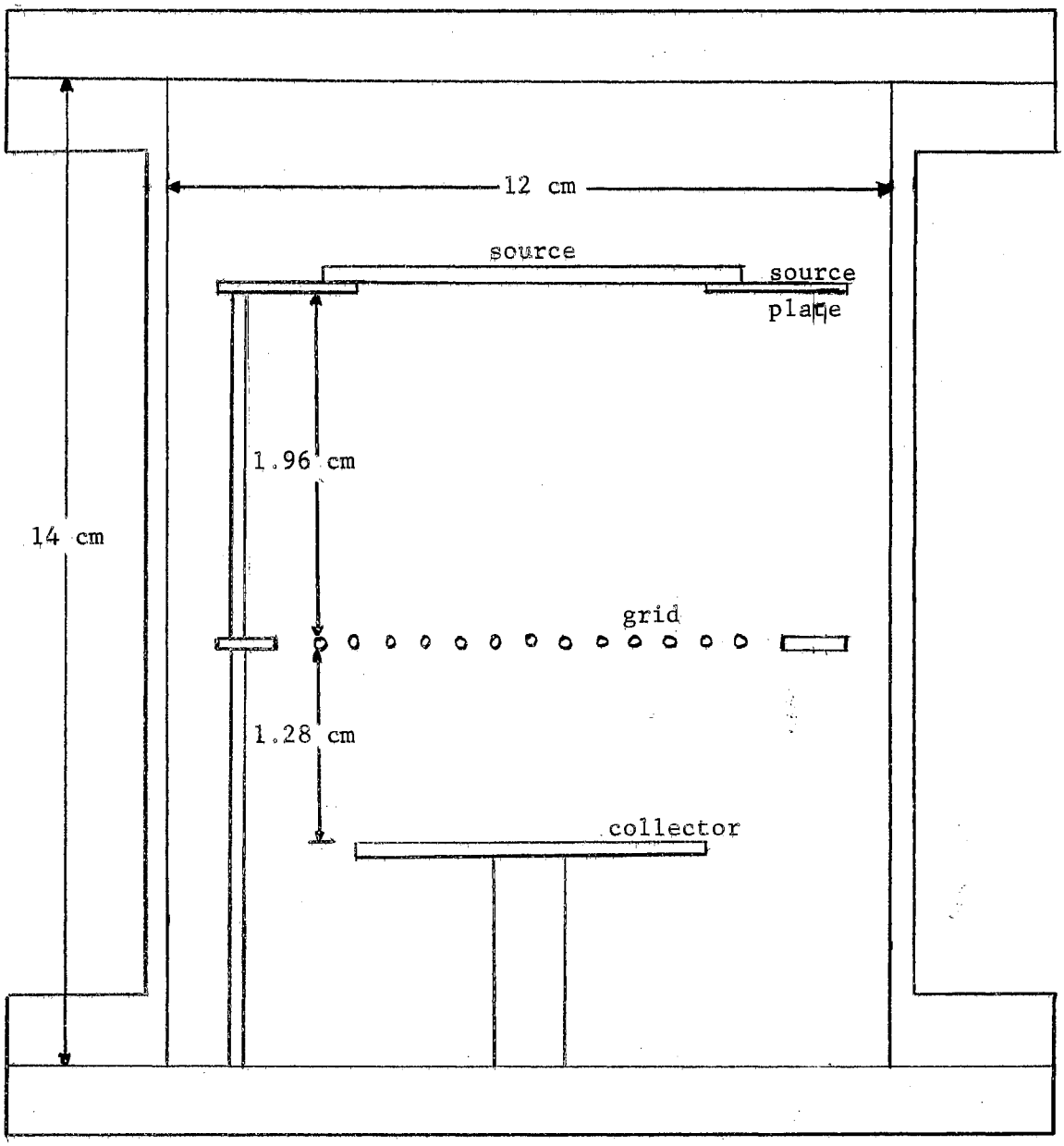

Figure 3. Cross Section of Ionization Chamber

When in use, the ionization chamber was filled to two atmospheres of a 90% argon-10% methane mixture. When an alpha particle was given off, it ionized the gas, forming a cloud of electrons. The electrons were then forced to the collector by the electric field. This negative pulse imparted to the collector was amplified by a Tennelec low noise preamplifier, model 100 A, which had been modified by Nurmia and described by Sievers (25). These pulses were then sent into a Nuclear Data pulse height analyzer. A block diagram of the apparatus is shown in Figure 4 below.

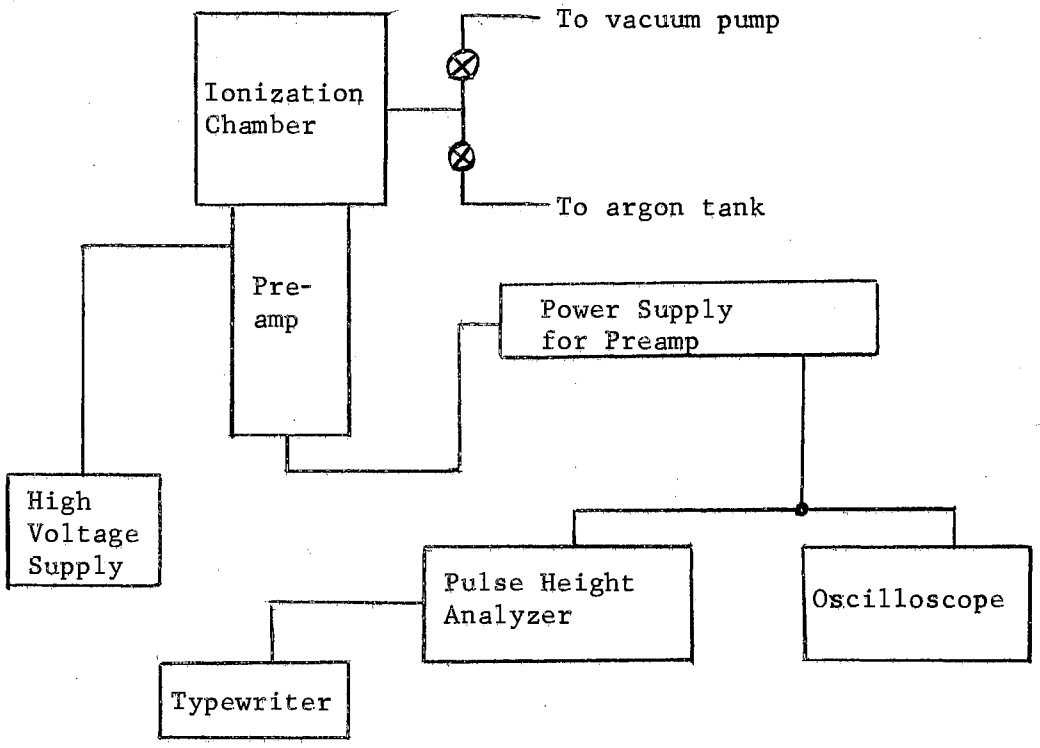

Figure 4. Block Diagram of Circuit

Figure 5 shows an actinium spectrum taken by the analyzer 14 hours after the source had been prepared. The determination of the various peaks in the spectrum was done in the standard way when a pulse height analyzer is used. The abscissa is measured in channel numbers going from 1 to 127 channels while the ordinate gives the corresponding number of counts in that channel. The channel to which any certain pulse entering the analyzer from the preamplifier falls, is determined by the size of the entering pulse. This pulse size in turn depends on the energy of the emitted alpha particle, for the higher the emission energy, the more ionization is produced and therefore a larger pulse enters the analyzer. The low energy pulses fall into the low channel numbers and as the pulse height increases so do the channel numbers in which the pulse is recorded. The analyzer is designed so that this energy dependence is linear. If the plot is linear, then the energy of each peak in the spectrum can be determined if two of the energies are known and their corresponding peaks identified. As shown in Figure 5, if an energy scale in mev is marked off on the right verticle axis and the two known energies are plotted in the channel numbers which have the peak or maximum number of counts in them, a straight line can be drawn between these two points. In this case the energy of Ac-227 (4.95 mev) and Po-215 (7.38 mev) are well known and these points were used to draw the line. The energy of any other peak is therefore readily found simply by finding the channel number that has the peak value and reading the corresponding energy from the straight line graph. Th-227 has a wide fine structure in that approximately 30% of the thorium atoms decay by a 5.75 mev alpha particle and 50% decay by a 6.0 mev alpha particle (8). The graph clearly shows this fine

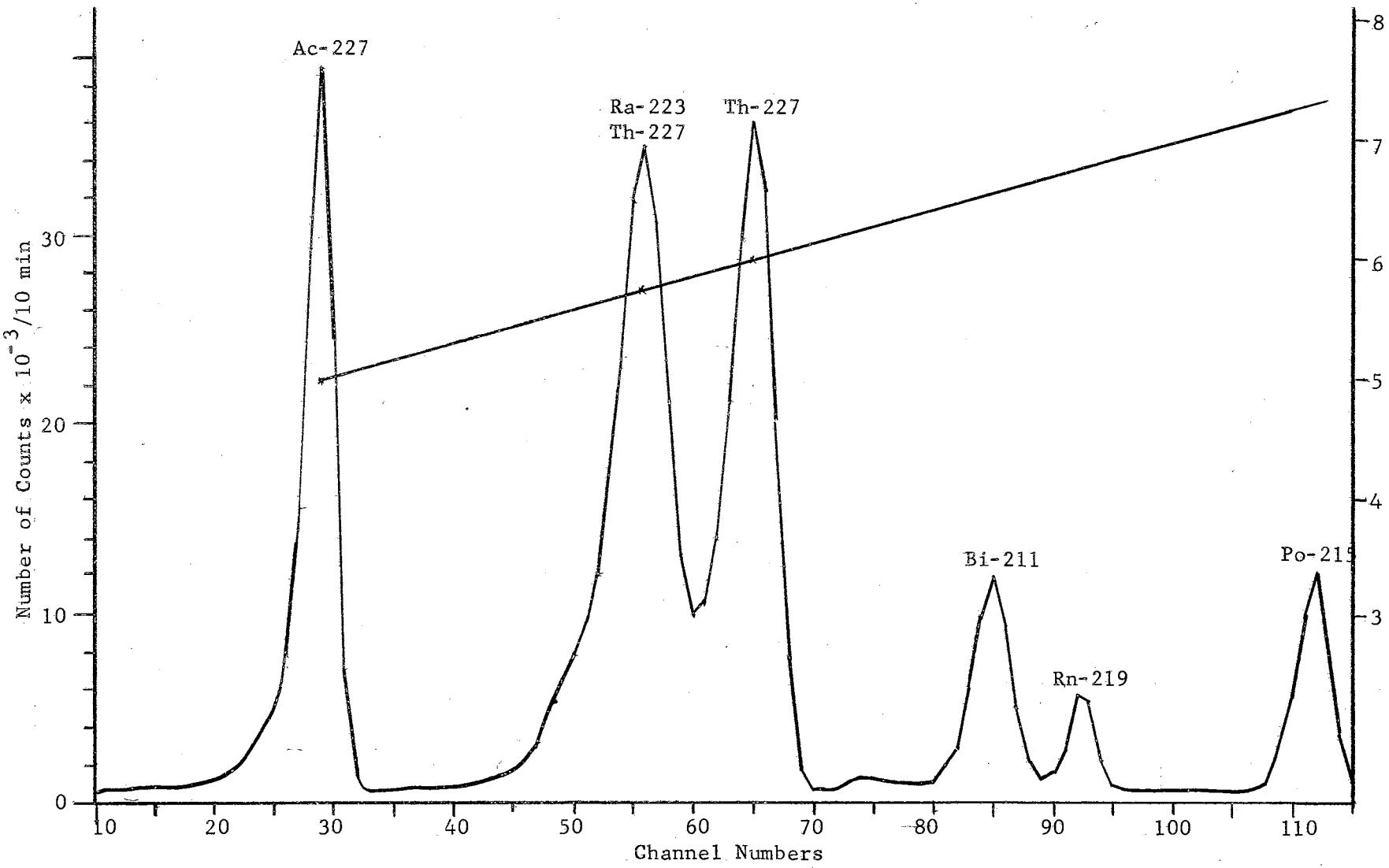

Figure 5. Spectrum Plot of the Actinium Series

structure and the energy agreement is excellent.

Preparation of Sources: Actinium

The original source of actinium used in this experiment was a five millicurie solution purchased from England. From the condition placed on Equation (6) only actinium is wanted in a source at $t = 0$. Therefore it was necessary to purify the actinium solution and get rid of the thorium and radium as well as the radium daughters. It was desired to remove at least 99.9% of the daughters such that the ratio of thorium counts/actinium counts is less than 0.1. A number of methods have previously been used to do such an extraction (26,27). For purposes here a solvent extraction using thenoyltrifluoroacetone (TTA) was done.

To give good statistics in a ten-minute counting period (10,000 counts) approximately 20 alpha particles per second need to be recorded by the analyzer. Since the chamber has a 50% geometry, a source giving approximately 40 alpha particles per second is required. This number divided by the branching ratio gives the strength of the original amount of source solution needed. Using the presently accepted value of the branching, the strength of the source would be

$$\frac{40}{.012} = 3300 \text{ d.p.s.} \approx .1 \text{ microcurie} \qquad (22)$$

This could be set as sort of a lower limit on the strength of a source that would still give good statistics as stronger sources than this were used.

Figure 6 shows the extraction curves for the members of the actinium series using TTA. From these curves it can be seen that if

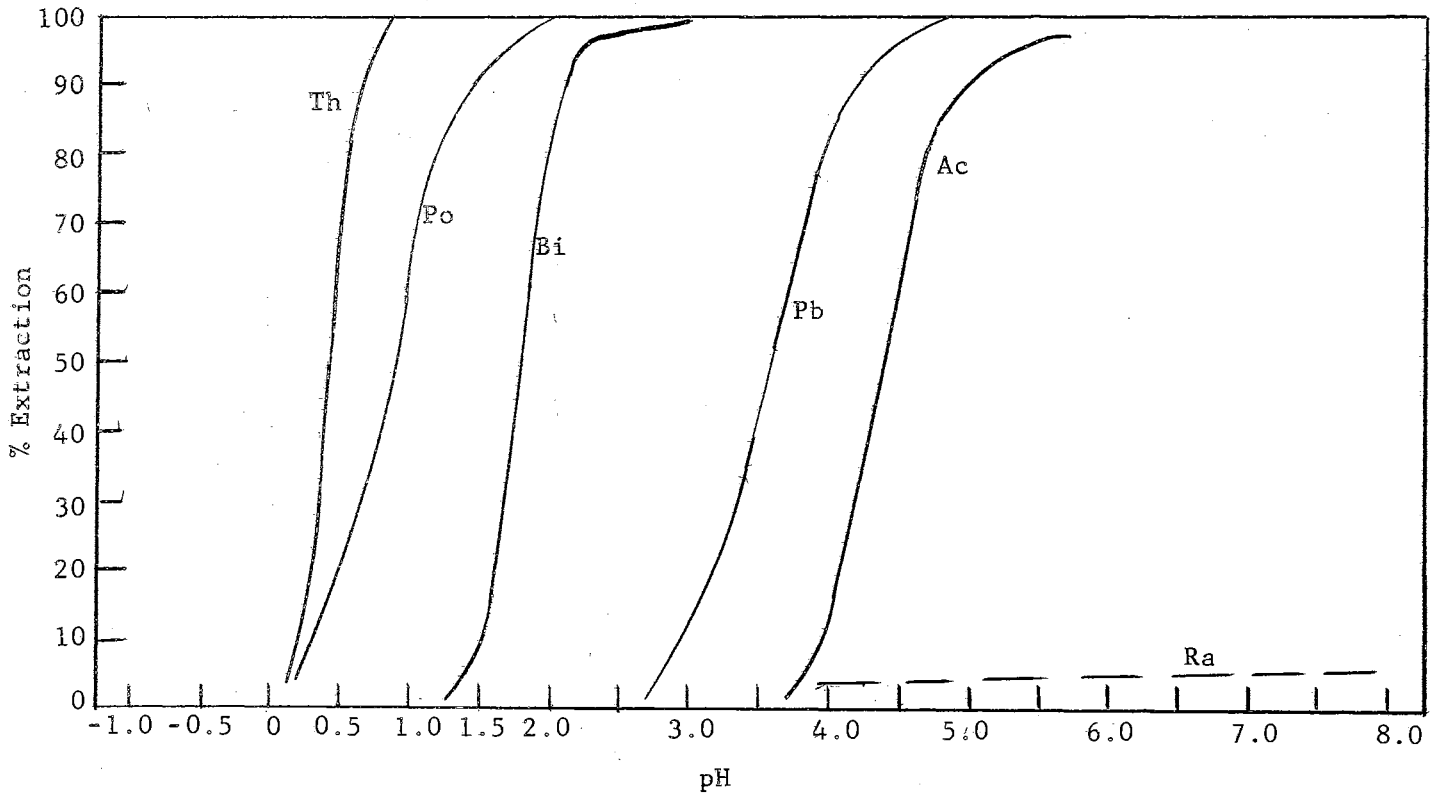

Figure 6. Extraction Curves for Actinium Series Using TTA

the source solution was raised to a pH of 5.7 and shaken with TTA in a separatory funnel, actinium and thorium would be extracted into the TTA while the radium would remain in the aqueous phase upon the separation of the two liquid layers. This step was performed using 0.4 M TTA and after the aqueous phase had been discarded the TTA was washed with an ammonium acetate-acetic acid 5.7 buffer to remove any remaining droplets of the source solution. Referring to the extraction curves again one can see that at pH 3.35, actinium will be removed from the TTA while the thorium will remain. Therefore to complete the extraction, a formic acid-ammonium hydroxide buffer of pH3.35 was added to the TTA and shaken in the separatory funnel. The aqueous phase could then be drained off and was the purified actinium solution. To make a source suitable for counting in the chamber, a few drops of solution were placed on a stainless steel planchet and slowly evaporated with a heat lamp. After evaporation, one drop of concentrated nitric acid was added to the warm planchet. This redissolved the source and allowed it to be spread evenly and thinly over a larger portion of the planchet. The heat of the planchet from the previous evaporation was sufficient to evaporate the acid. The source was then flamed to remove any volatile salts or traces of moisture. This, then, was the source that was put in the chamber.

The above mentioned procedure of preparing sources was used for about one-third of the runs made to determine the branching. For the rest of the sources a one-hour waiting time was used between the time when the final purified Ac-227 solution was prepared and the placing of the solution on the planchet for counting. Looking at Figure 5 one can see that at pH 5.7, lead is carried along with the actinium and

goes into the TTA. When the 3.35 buffer is added, some of the lead again comes along with the actinium into the final source solution. Since lead is one of the daughters of radium and radium and thorium have been removed to about 99.9%, the lead will die out with a 36.1 minute half life (28). The lead is a beta emitter and does not itself bother counting with the ionization chamber. However the lead decays to Bi-211 which is an alpha emitter and so it was desirable to remove this peak. In one hour the number of counts in the bismuth peak had become much less than the number of counts in the actinium peak and so the bismuth had effectively been removed. The runs made with the lead and bismuth were not invalid, however, since all needed measurements could still be performed but with increased difficulty and calculation time. In these runs where the bismuth peak was present, the bismuth tail extended under the actinium peak and therefore had to be subtracted. The first runs were the most important ones in determining the actinium alpha particles per second since no radium or thorium tail was yet present. For these reasons it was desirable to remove this bismuth contribution leaving the actinium peak alone. Figures 7 and 8 show a comparison of these two cases.

A freshly prepared source was placed in the chamber and the air was removed by a vacuum pump and the chamber then filled with two atmospheres of the argon-methane mixture. The strengths of the sources used in the experiment were varied over a wide range going from as low as 2.56 counts per second to as high as 166 counts per second. This was done to make sure the branching did not vary as the strength of the source changed. Eighty-minute runs had to be used for the weak sources to keep good statistics. For strong sources the analyzer was

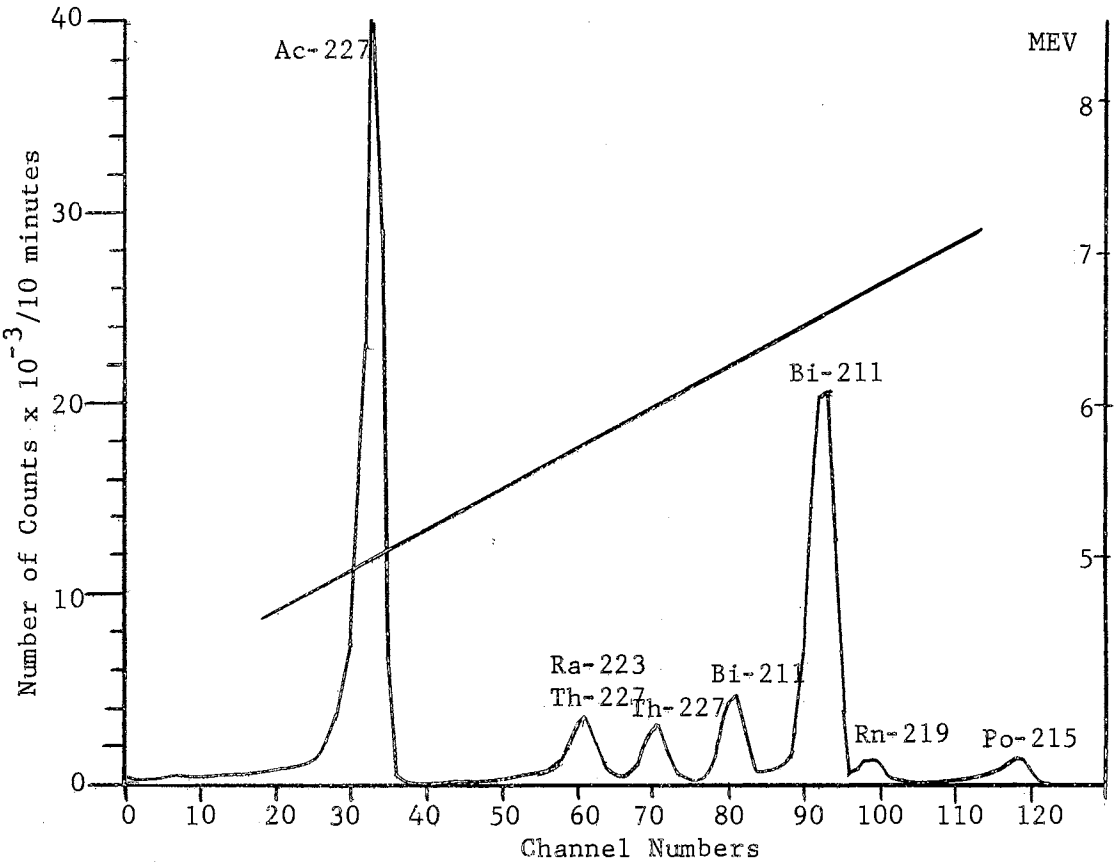

Figure 7. Spectrum Plot of a Freshly Prepared Source with Bi-211

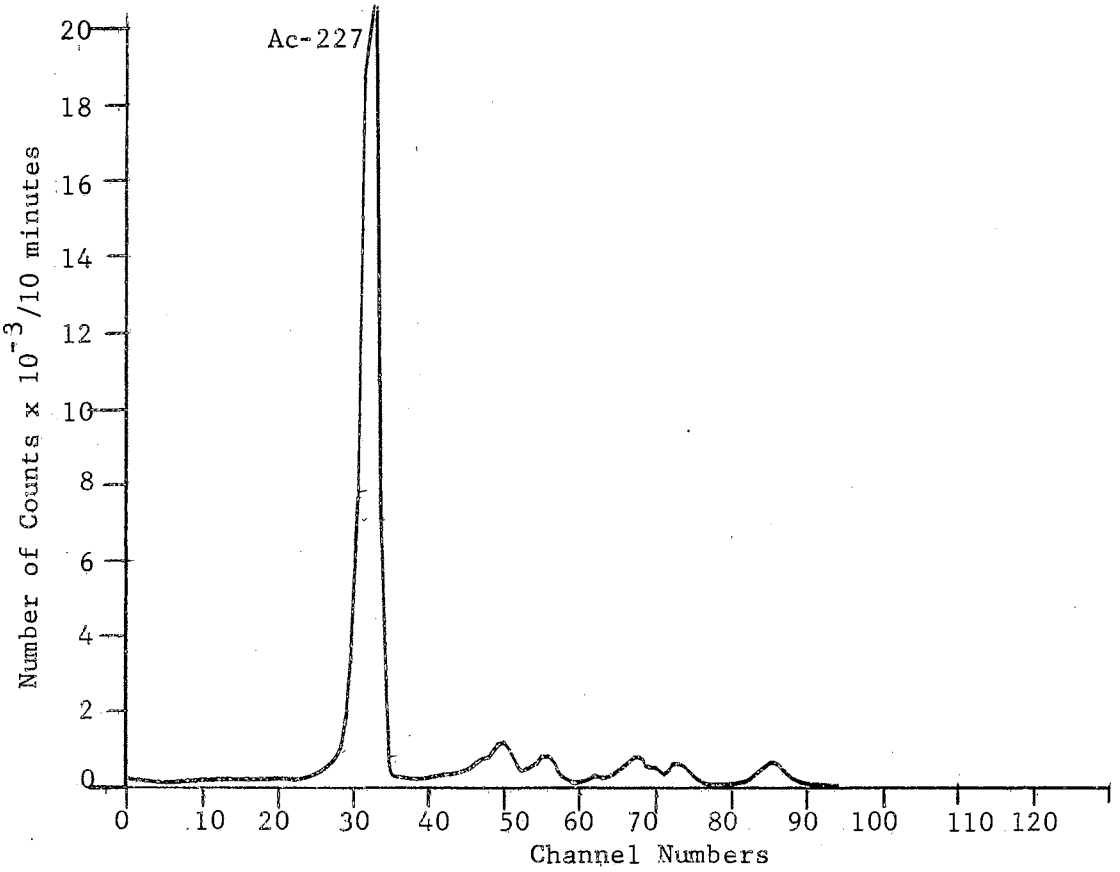

Figure 8. Spectrum Plot of a Freshly Prepared Source with Bi-211 Decayed

put on ten-minute automatic repeat runs, so that at the end of each ten minutes the contents of the memory were automatically typed out by the typewriter, the memory erased, and a new run begun.

Preparation of Sources: Francium

The purification of francium from all other radioactive series members follows closely to the procedure used in the actinium separation. The original source solution which contained actinium, thorium, francium, and radium was raised to pH 5.7 by the addition of NH_4OH. This solution was then shaken in a separatory funnel with a like amount of 0.4 M TTA. As before the actinium and thorium were extracted into the TTA while the radium and francium remained in the aqueous phase. This phase was drained off leaving only the TTA. The TTA solution was washed several times with the ammonium acetate-acetic acid 5.7 buffer to remove any remaining droplets of radium. The TTA containing the thorium and actinium was allowed to stand for one hour. The half life of francium is approximately 1/3 of an hour and so in an hour, three half lifes have passed and the francium has grown into 87½% of its equilibrium value. If some 5.7 buffer is added to the TTA in the separatory funnel and the solution is shaken, the actinium and thorium remain in the TTA while the francium separates into the aqueous phase as can be seen again from the curves in Figure 6. Even though the actinium and thorium have been removed, this aqueous source solution still contains lead and bismuth along with the francium. An extraction with dithizone was used to make this separation. Figure 9 shows the extraction curves of lead and bismuth using dithizone. If the source solution was raised to a pH of 8.5, it can be seen from the curves of

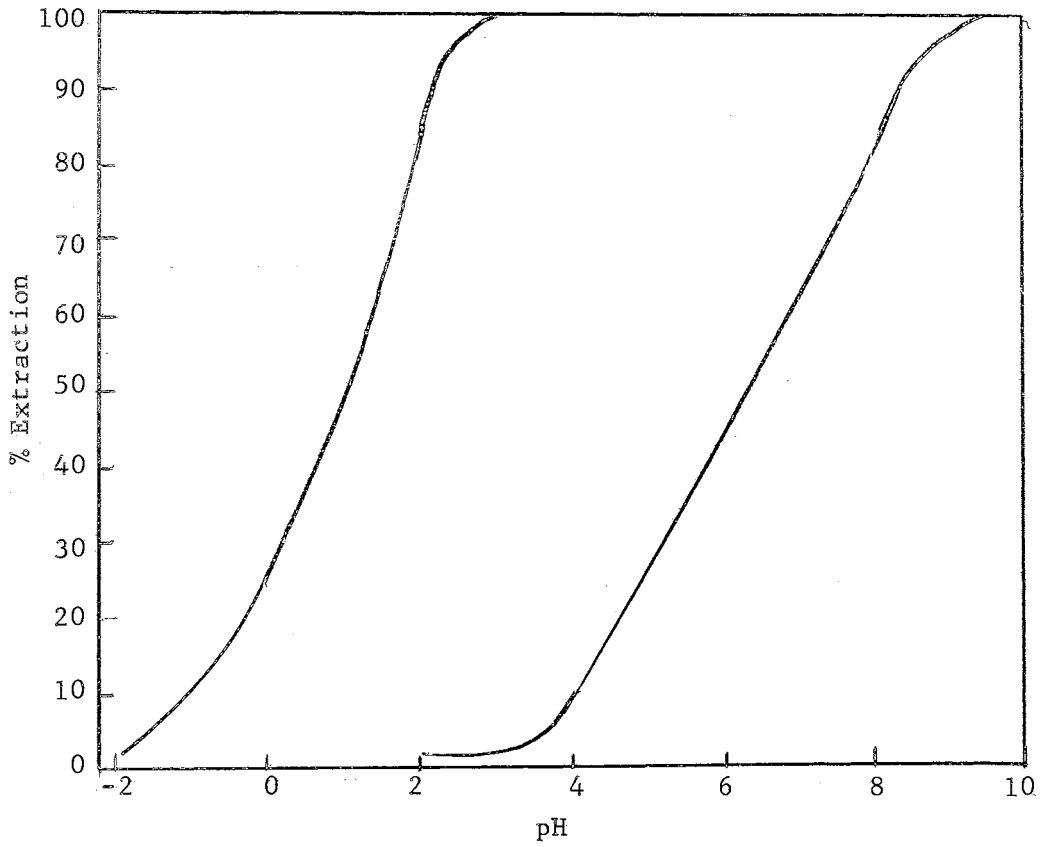

Figure 9. Extraction Curves of Lead and Bismuth Using Dithizone

Figure 9 that both the lead and bismuth will be extracted into the dithizone. The source solution was raised to pH 8.5 by the addition of NH_4OH. One milligram of lead nitrate was added to the solution as lead carrier. To the separatory funnel was then added the dithizone and the mixture shaken for several minutes. In this extraction there is a good check to see if all the lead and bismuth are in the dithizone, for if there is no free dithizone in the funnel, the dithizone turns a bright cherry color. It is only necessary to add more dithizone to the funnel until some of the original blue color of the dithizone remains. The blue color indicates free or unused dithizone and assures a good extraction. After the two liquid layers had separated, the lower dithizone phase was drained off leaving the francium source solution. This solution was placed at the top of a cation exchange column. The column consisted of 200-400 mesh Dowex 50 (hydrogen form) contained in a glass tube. The tube had inside diameters of 2 mm for 8 cm of length and 7 mm for 9 cm of length. The constricted portion of the tube contained the resin while the larger served as the reservoir. The column was jacketed and pressure was supplied by a nitrogen tank connected to a mercury column. After the feed had passed, the column was eluted with 1 N HNO_3. With the jacketed column at a temperature of 70° C and the pressure at 20 mm of Hg, 4 drops per minute were obtained from the column. If any impurities existed in the source solution before it was added to the column, they were held in column longer than the francium, the francium being the first element to pass. The source drops were caught on a stainless steel planchet and evaporated under a heat lamp. The planchet was first counted in a flow counter to see if there was any alpha activity. If some alpha activity existed

the source was no good as some radium or thorium still contaminated the source. If no alpha activity was present, the beta activity was measured with the G-M tube and the half life determined.

CHAPTER III

RESULTS

Actinium Branching

A typical actinium source was counted for a 24 to 48 hour counting period. As shown in Figure 10, the actinium peak has a constant value in a ten-minute timing period where these ten-minute runs were made at the start of the source and also after three hours, twenty hours, and fifty hours had passed. This superposition of the actinium spectrum at these various times also shows the way in which the thorium grows in as well as the growth of the radium and the radium daughters. The slight rise in the actinium peak is due to the thorium-radium tail that grows in under the actinium peak. For the actual calculation it was necessary to total the counts in the individual peaks. The output data from the analyzer was typed out automatically channel by channel and an example of this output is shown in Figure 11. From this data one could plot the actinium spectrum as it existed at that particular time. From such a graph one can determine the channels that make up the peak and these can then be marked off on the output data (see Figure 11). Since the number of channels making up the peak reamined the same it was necessary to only mark off the appropriate channels on each successive piece of output data and it was not necessary to plot each ten-minute run. During the total timing period it sometimes happened that peaks did shift down in channel numbers as they did in

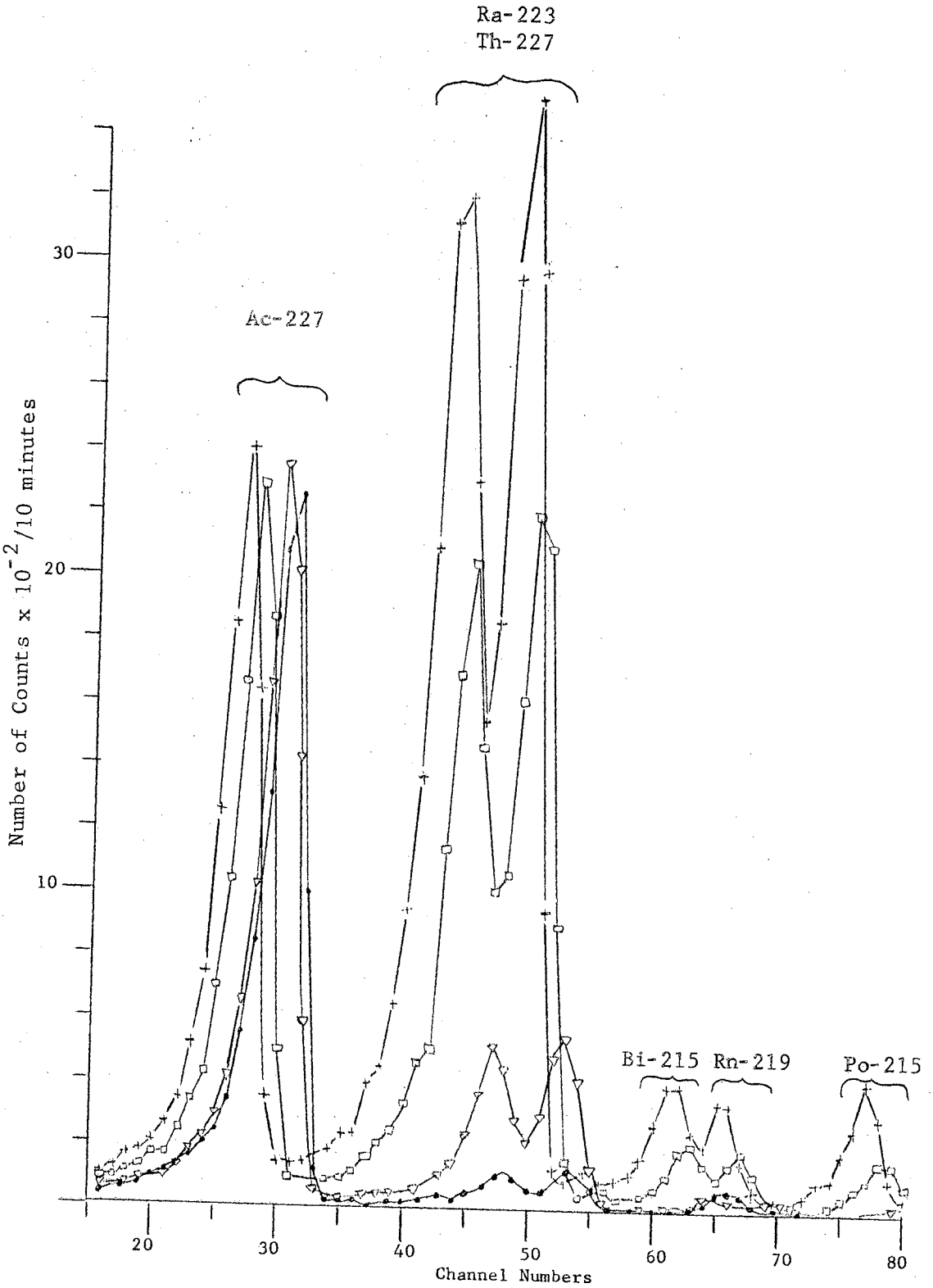

Figure 10. Superposition of Actinium Spectrum at Various Times after Separation

```
100000 000101 000070 000070 000053 000055 000057 000067
 000060 000070 000076 000074 000087 000081 000097 000099
 000099 000116 000122 000129 000138 000156 000174 000208  Ac
 000263 000346 000418 000623 000970 001437 002103 002398
 001096 000266 000181 000169 000202 000202 000269 000268  Th
 000381 000411 000584 000809 001191 001784 002931 004101
 004117 002928 001980 002497 003821 004839 003977 001468
 000245 000109 000146 000163 000150 000133 000166 000220
 000394 000552 000538 000410 000323 000481 000508 000292
 000132 000050 000029 000029 000028 000043 000104 000193
 000391 000552 000490 000353 000121 000031 000012 000007  Po
 000004 000004 000000 000000 000001 000000 000001 000000
 000000 000000 000001 000002 000000 000000 000000 000000
 000000 000002 000000 000001 000001 000000 000000 000001
 000000 000000 000000 000000 000000 000001 000001 000000
 000000 000001 000000 000001 000001 000001 000000 000002
100000 000112 000079 000080 000069 000058 000060 000056
 000082 000070 000056 000072 000067 000075 000085 000092
 000092 000088 000103 000134 000115 000160 000200 000200  Ac
 000242 000359 000445 000649 000911 001529 002193 002350
 001049 000224 000145 000187 000190 000234 000223 000288  Th
 000360 000439 000617 000777 001213 001990 002956 004206
 004190 002730 002035 002607 004047 004877 003684 001348
 000218 000114 000130 000179 000150 000125 000173 000267
 000399 000578 000583 000401 000335 000444 000510 000295
 000128 000038 000022 000041 000049 000058 000103 000209
 000430 000519 000493 000278 000123 000023 000012 000005  Po
 000007 000002 000001 000000 000001 000000 000001 000001
 000000 000001 000001 000001 000001 000000 000002 000002
 000001 000001 000000 000000 000000 000001 000001 000000
 000000 000000 000000 000001 000000 000000 000000 000000
 000000 000001 000000 000000 000001 000001 000001 000000
100000 000093 000076 000069 000077 000067 000066 000078
 000055 000063 000064 000069 000068 000080 000077 000072
 000087 000103 000111 000139 000142 000152 000175 000209  Ac
 000245 000331 000452 000590 000922 001522 002150 002391
 001124 000259 000156 000163 000182 000212 000242 000325  Th
 000343 000446 000613 000780 001204 001921 003037 004141
 004074 002834 001962 002606 003895 004892 003843 001451
 000237 000122 000153 000168 000163 000134 000175 000247
 000394 000615 000616 000383 000355 000484 000548 000318
 000109 000043 000030 000027 000040 000058 000096 000221
 000404 000555 000536 000356 000123 000032 000009 000007  Po
 000001 000002 000001 000000 000000 000000 000000 000001
 000000 000003 000002 000000 000000 000002 000000 000000
 000000 000000 000000 000001 000001 000000 000000 000001
 000000 000000 000001 000001 000000 000000 000000 000003
 000002 000000 000001 000000 000000 000001 000000 000000
```

Figure 11. Sample of Output Data

Figure 10. However in such a shift the width of the peak remained the same. To make sure this was accounted for in the summing of the peaks, a plot of the spectrum was made at various intervals in a run and if a shift had occurred, the appropriate channels were used. Typical widths for the actinium and thorium were 10 and 16 channels respectively. The thorium peak was wider because its alpha spectrum had a fine strucutre with two main groups being 250 Kev apart. An energy spread of this much is detectable by the analyzer.

From Figure 10 of the superposition of the spectrum at various times, two additional things may be pointed out. The first is that there was a general background level present which had to be subtracted, and secondly the tail from the thorium-radium peak extended under the actinium peak. As the time increased and the counts in the thorium peak increased, the tail also increased and caused a rise in the actinium peak. These additional counts also had to be subtracted.

The general background level was low and did not present a serious problem. A look at the curve from an early run in Figure 10 will show that the count rate after the actinium peak drops off very sharply to a minimum. In the ideal case this would go all the way to zero since it is not possible for the Ac-227 alpha particles to produce any pulses in these channels. At the same time there would be no tail contribution from the thorium and radium since at the beginning of the run they are essentially removed. The average amount of counts, therefoure, over a number of early runs in these channels just following the actinium peak will give the general background level. This will be the correct level for the entire run as the background cannot increase.

As could be expected, the background level under the thorium peak was not as great as the background level under the actinium peak since the channels corresponding to the thorium peak are higher in energy. In fact the background was one plateau under the actinium peak and another plateau under the thorium peak, the drop from one plateau to the other fortunately occurring between the peaks. The same arguments used to determine the background under the actinium peak can be used on the thorium peak. The count rate from the thorium peak to those channels just following it drops off very rapidly. The average of the counts for early runs in these channels just following the peak give the background level for thorium. This again is due to the fact that no thorium alpha emissions could produce counts in these channels of higher energy and there is yet no tail contribution in these channels from the radium daughters, all of whose peaks are of higher energy and will later give a tail contribution.

The tail of any peak is caused by alpha particles which have lost some of their initial energy. The main way a particle loses energy is due to absorption in a thick source. In such a source the particle makes a number of collisions before leaving the source and consequently causes less ionization in the gas due to its lower energy. This along with back scattering produces a tail from the highest energy given off back to zero energy.

Since the presence of this tail from the high energy peak gives a greater number of counts to the lower energy peak than really belong, this contribution must be subtracted. On a graph, the tail can be estimated and the proper amount to subtract can be found. Figure 12 shows such a plot. However to do this for every run in a 24 to 48 hour

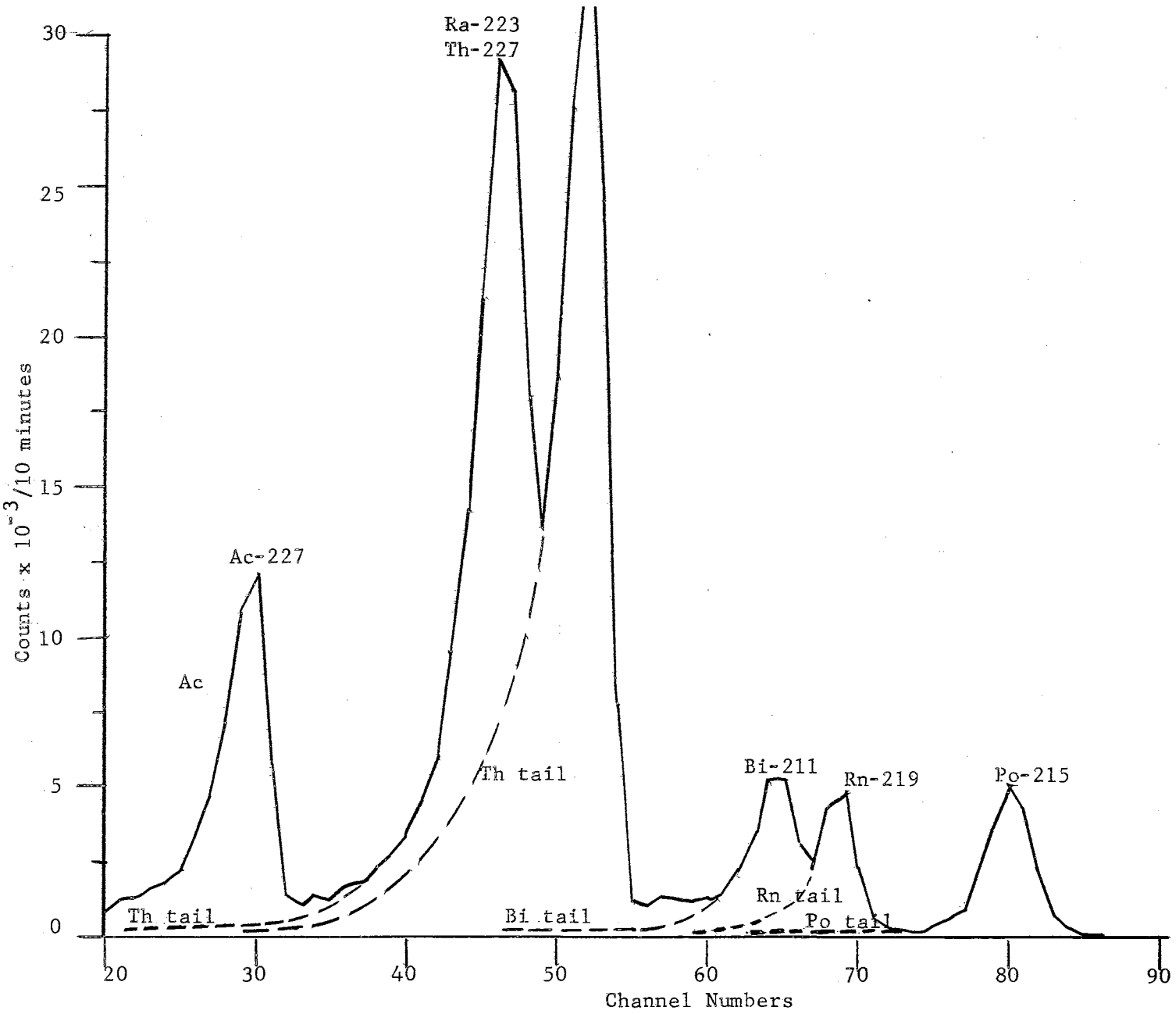

Figure 12. Actinium Spectrum Showing Tail Contributions

period would have been impracticable, and therefore only a few plots were made at times near the start, middle and end of the run. It was then possible to interpolate between these values to get the correct value to subtract.

There was one other contribution in the thorium peak that had to be subtracted. As shown in Figure 1, thorium has a double peak at 6.0 mev and 5.75 mev while radium has an alpha energy of 5.7 mev and therefore lies under the lower thorium peak. This extra contribution will give a wrong growth rate for thorium and therefore had to be subtracted. This however could be done quite easily by calculating the number of counts in the Po-215 peak since Po-215 is in equilibrium with the radium. Another look at Figure 1 will show that Rn-219, the daughter of Ra-223, has a half life of 3.92 seconds (29) and decays into Po-215 which has a half life of 1.83 milliseconds (30). Therefore as soon as a radium alpha particle is emitted, it is promptly followed by alpha particles from radon and polonium. The alpha particle emitted from polonium has an energy of 7.38 mev (31) and is a peak standing alone (see Figure 5). Thus the number of counts in this peak can be added up and the sum subtracted from the number of counts in the thorium peak and the radium contribution has been removed.

With these corrections made, the number of counts in the thorium peak at successive times were known. The increasing count rate was then plotted against the reduced time, t', so the slope could be found. Since a least squares fit of data is desirable, the slope was found using a linear regression deck on an IBM 650 computer. The final listing gave the points on the curve, the error for each point, and the slope and its error. Figure 13 shows a typical thorium growth rate plot.

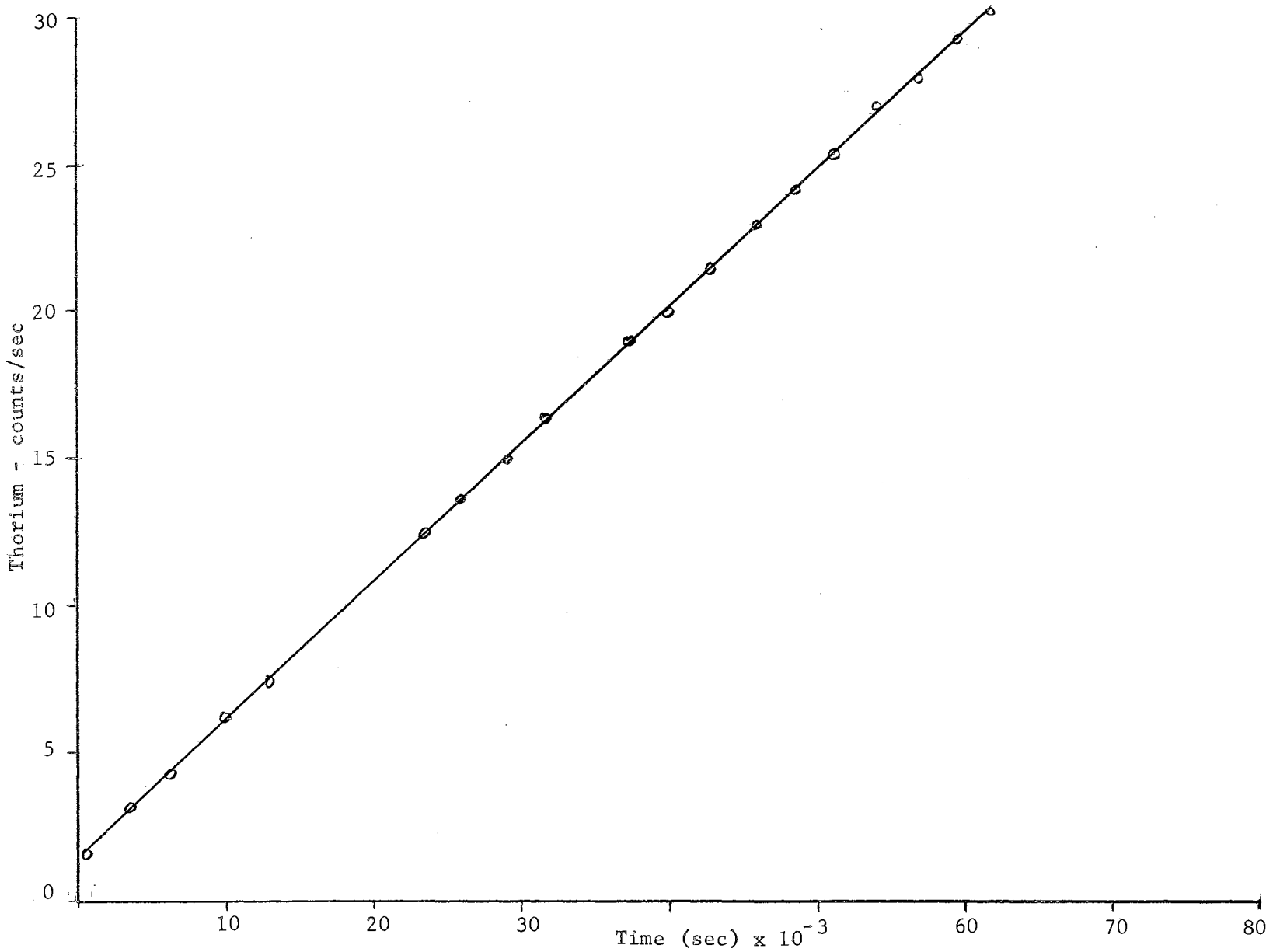

Figure 13. Thorium Growth Rate Curve

With these values of A_1 and dA_3/dt' now obtained the ratio of the partial decay constants can be found from Equation (21) and the branching ratio determined from Equation (2). Table I gives the number of actinium alpha particles observed per second, the growth rate of thorium, and the branching ratio for the various samples made. A sample calculation of a branching measurement is given in Appendix A including all the various corrections that had to be made.

TABLE I

ACTINIUM BRANCHING RATIO RESULTS

Source	Number of Actinium Alpha's per Second	Growth Rate of Thorium counts/sec^2	Branching Ratio
1	2.705 ± .023	[8.227 ± .164] x 10^{-5}	1.430 ± .031 %
2	15.609 ± .161	[4.689 ± .013] x 10^{-4}	1.447 ± .016 %
3	17.553 ± .171	[5.168 ± .011] x 10^{-4}	1.476 ± .015 %
4	7.962 ± .115	[2.319 ± .026] x 10^{-4}	1.491 ± .021 %
5.	106.88 ± .422	[3.149 ± .008] x 10^{-3}	1.475 ± .007 %
6	165.88 ± .526	[4.995 ± .036] x 10^{-3}	1.441 ± .006 %
7	144.48 ± .491	[4.549 ± .011] x 10^{-3}	1.381 ± .006 %

Francium Half Life

Three Fr-223 sources were prepared. The decay by beta emission was counted with a G-M counter and the half life determined. This was done by making a least squares fit of the data on the IBM 650. The data was weighted as N^2/N_o where $N = N_o$ - background, and N_o was the observed counts. Table II shows the three values obtained for the half life and the associated error.

TABLE II

FRANCIUM HALF LIFE RESULTS

Source	Half Life	Decay Constant
1	21.67 ± .17 min	0.032 min^{-1}
2	22.57 ± .14 min	0.031 min^{-1}
3	22.99 ± .16 min	0.030 min^{-1}

CHAPTER IV

DISCUSSION

Actinium

The value of the branching ratio commonly accepted at the present time is 1.25%. From Table I, the mean of the branching ratios measured in this work was 1.45 ± .06%. The error in the branching ratios given in Table I came from the two measured quantities A_1 and dA_3/dt'. The major contribution to this error was in A_1, the observed actinium counts per second, as consistancy in the number of counts in the actinium could never be obtained closer than about 3%. On the other hand the rate of growth of thorium could be measured with much better accuracy, the error in the slope being as low as 0.2% in one case. However the calculations used to figure the branching ratio for the various sources gave error limits that did not include all the other values of the branching ratio from the other sources. This indicates that there was an error present that was not included in the calculation and therefore the ± .06 value was given as the error limit since the branching ratio surely seems to lie in this range of 1.45 ± .06%.

Using this value of the branching ratio, the partial half life for actinium alpha emission is the total half life, 22 years, divided by the branching ratio.

$$T_{\frac{1}{2}(\alpha)} = \frac{22 \text{ years}}{.0145} = 1520 \text{ years}$$

A similar calculation using the 1.25% branching gives a partial half life of 1760 years. An interesting comparison is a plot of the log of experimental partial half lives against a calculated parameter given by the equation

$$X = \left[\frac{(Z - 2)}{\sqrt{E}} - (Z - 2)^{2/3} \right]$$

where Z is the atomic number of the parent element and E is the alpha particle's energy (see Figure 14).

The straight line on the plot was drawn through points corresponding to U-238, Th-232, Ra-226, and Rn-219 (32). The formula used was

$$\log T = C_1 X - C_2 \qquad (25)$$

where X is given in Equation (24). The other nuclides used in this figure are those with N (number of neutrons) equal to 138 and which are alpha active, and members of the main actinium series. The log of the partial half lives for Ac-227 figured with the two branching ratios are both shown on the graph.

The new value of the partial half life gives a partial decay constant of 4.48×10^{-4}/years. This decay constant can be compared with the value obtained from theoretical considerations of barrier penetration. Kaplan (22) gives an expression for the disintegration constant in an approximate form as

$$\lambda = \frac{v_\alpha}{r_o} \exp \left[- \frac{8(Z - 2)e^2}{\hbar v_\alpha} (\alpha_o - \sin \alpha_o \cos \alpha_o) \right] \qquad (26)$$

where $\alpha_o = \text{avc cos} \left[\frac{M v_\alpha^2 r_o}{4e^2 (Z - 2)} \right]$ is the effective radius of the nucleus for alpha decay; v_α is the velocity of the alpha particle relative to the nucleus, and is equal to $v(1 + \frac{M}{M_r})$ and where v is the alpha particle's measured velocity and M_r is the mass of the recoil

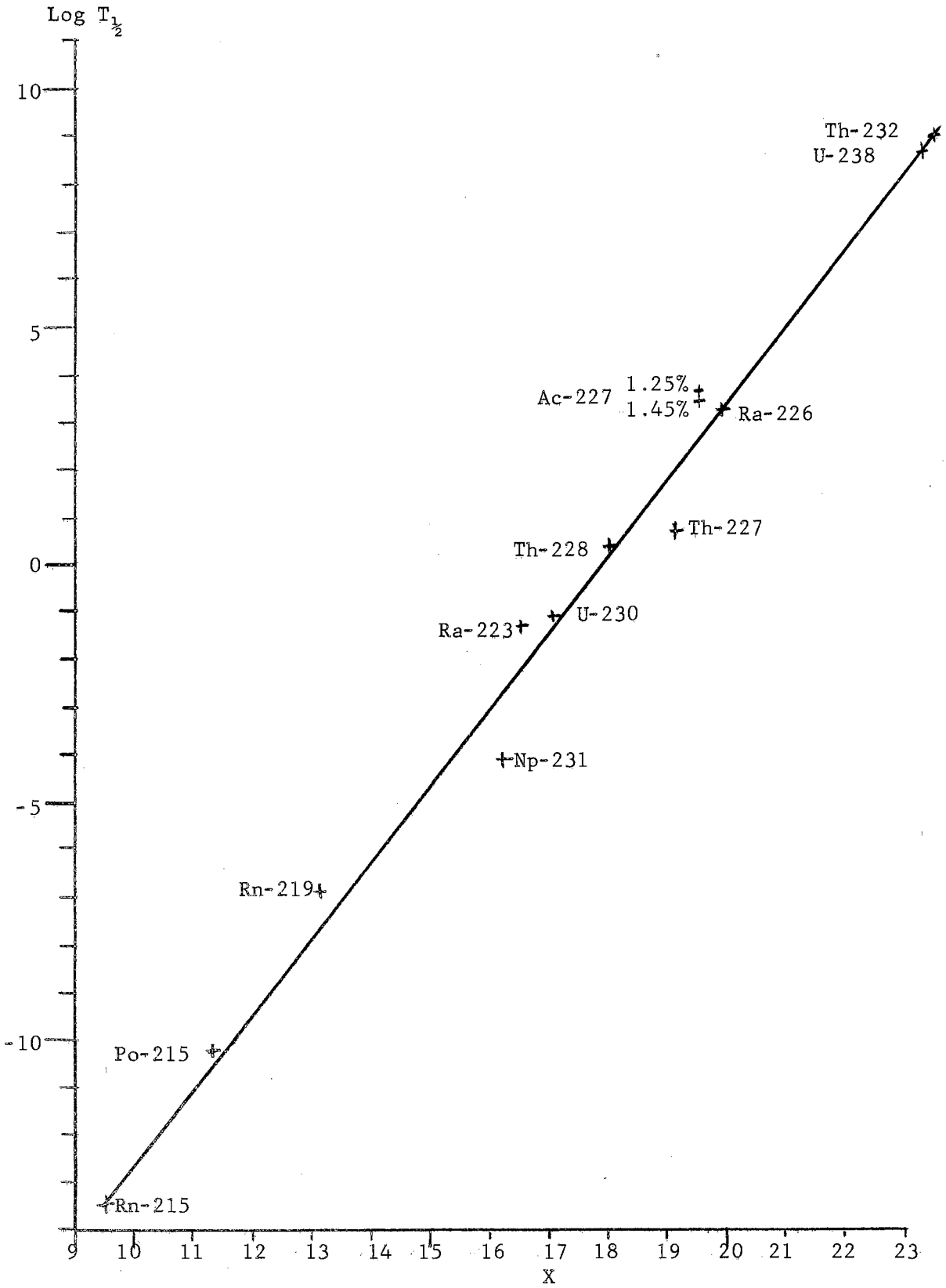

Figure 14. Comparison of Experimental Partial Half Lives

nucleus. The radius used in the formula was $1.5 \times 10^{-13} A^{1/3}$ cm and the velocity was found by using the emission energy of the alpha particle from actinium. The partial decay constant found in this manner had a value of 0.158/year which leads to a half life of 4.39 years, where as the experimental value is 1520 years. This longer experimental half life however is what one should expect. The theoretical value is less because first the theory assumes that there is always an alpha particle formed in the nucleus and constantly trying to get out. Secondly, the theory assumes an alpha particle will always have all the energy available which also is not the case.

Francium

As mentioned in the introduction, it was felt that the presently accepted value of 22 ± 1 minutes could have been obtained if, in the preparation of the source from which this measurement was made, some lead contamination was present. If this were the case, the value of the half life that was measured would have been too large since the half life of Pb-211 is 36 minutes. Twenty-two minutes would then be a possible value between 17 minutes and 36 minutes. As Table II shows, however, the values obtained here agree generally with the presently accepted value while the sources here used were checked against Pb-211 contamination by measuring their alpha activity first. The three values here obtained for the half life were not in close enough agreement to give any improvement on the 22 ± 1 minute value, but they do give another measurement generally confirming the previous measurement. More accurate measurements could be obtained, most likely, if more active source solutions were used at the beginning of the chemical separation

since the low count rate was a limiting factor in computing the half life.

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APPENDIX A

The following table shows some typical values of the various quantities used in making a branching measurement. A number of ten-minute runs at various times were chosen to show the increase in thorium counts, the increase in tail contribution, and the radium contribution to the thorium peak.

TABLE III

SAMPLE CALCULATION

Run	1	16	28	52	100
Number of Counts in Ten Minutes in Ac Peak	9,661	9,911	10,080	---	---
Number of Counts in Ten Minutes in Th Peak	1,247	4,821	9,543	15,504	25,654
Number of Counts in Ten Minutes in Po Peak	117	208	354	602	1,268
Background and Tail Counts to be Subtracted from Ac	360	450	720	---	---
Background and Tail Counts to be Subtracted from Th	218	308	554	902	1,968
Corrected Ac cts	9,301	9,461	9,360	---	---
Corrected Th cts	1,030	4,513	8,989	14,602	23,686
Time (sec)	750	12,904	29,012	49,000	83,290

The corrected thorium counts per second and the time were punched on data cards for the IBM 650. A fortran deck was first used to calculate the reduced time t' from t, and then a least squares fit was made of the data. The slope of the curve, i.e. the rate of growth of throium for this example, was $[4.689 \pm .0147] \times 10^{-4}$. The average number of actinium counts per second was 15.609 cps. Then from Equation (21) the ratio of the partial half lives is

$$\frac{\lambda_{1\alpha}}{\lambda_{1\beta}} = \frac{\lambda_2 A_1}{dA_3/dt'} = \frac{[(4.31) \times 10^{-7}][15.609]}{[4.689 \times 10^{-4}]}$$

$$\frac{\lambda_{1\alpha}}{\lambda_{1\beta}} = 1.468 \times 10^{-2}$$

Equation (2) can now be used to find the branching ratio

$$\text{Branching ratio} = \frac{\lambda_{1\alpha}/\lambda_{1\beta}}{1 + \lambda_{1\alpha}/\lambda_{1\beta}} = \frac{1.468 \times 10^{-2} \times 100}{1 + 1.468 \times 10^{-2}}$$

$$\text{Branching ratio} = 1.447\ \%$$

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