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THE COMPLEX ALPHA SPECTRA OF THE HEAVY ELEMENTS

Francesco Asaro

(Thesis)

June, 1953

Berkeley, California

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THE COMPLEX ALPHA SPECTRA OF THE HEAVY ELEMENTS

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June, 1953

ABSTRACT

Using a 75 cm radius of curvature 60° symmetrical electromagnetic analyzer, we have investigated the complexity of the alpha spectra of $Cm^{244, 243, 242}$, $Am^{243, 241}$, $Pu^{242, 241, 240, 239, 238}$, $U^{234, 233, 232, 230}$, $Th^{228, 226}$, $Ra^{226, 224, 223, 222}$, $Em^{212, 211, 210, 208}$, $At^{211, 210}$ and $Po^{211, 209, 208}$. We have also investigated the gamma rays of $Cm^{243, 242}$, Am^{243}, Pu^{238}, U^{234}, Th^{228} and Em^{211}.

Decay schemes have been proposed for many of the nuclides and empirical correlations have been determined for the energies and intensities of the alpha groups observed in the even-even nuclei. These correlations have been evaluated with respect to alpha decay theory and recent developments in the literature.

THE COMPLEX ALPHA SPECTRA OF THE HEAVY ELEMENTS

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I. INTRODUCTION

A study of complex alpha energy spectra was undertaken in the hope that a correlation with alpha systematics might promote a better understanding of nuclear states and their influence on the alpha decay process.

II. EXPERIMENTAL APPARATUS AND TECHNIQUES

A. Alpha Particle Spectrograph

The alpha particle spectrograph used in the present measurements is a converted Nier type mass spectrograph. It employs a 60° symmetrical magnetic analyzer and the normal trajectory has a radius of curvature of 75 cm. Fig. 1 is a schematic diagram of the optics of the instrument and shows the magnet, source, slit system and detector. The magnetic field is produced by an electromagnet with a 1 in. gap between the pole pieces and at maximum intensity is capable of bending a 14 Mev alpha particle. The entire system of source, magnetic gap and photographic plate is evacuated to a pressure of 10^{-4} to 10^{-6} mm Hg prior to operation.

A detailed description of the instrument, its power supply and operating characteristics has been published elsewhere.[1] A brief summary of the characteristics is given here. The magnet power supply is capable of maintaining the current constant to one part in 10,000 over at least a 24-hour period. The current

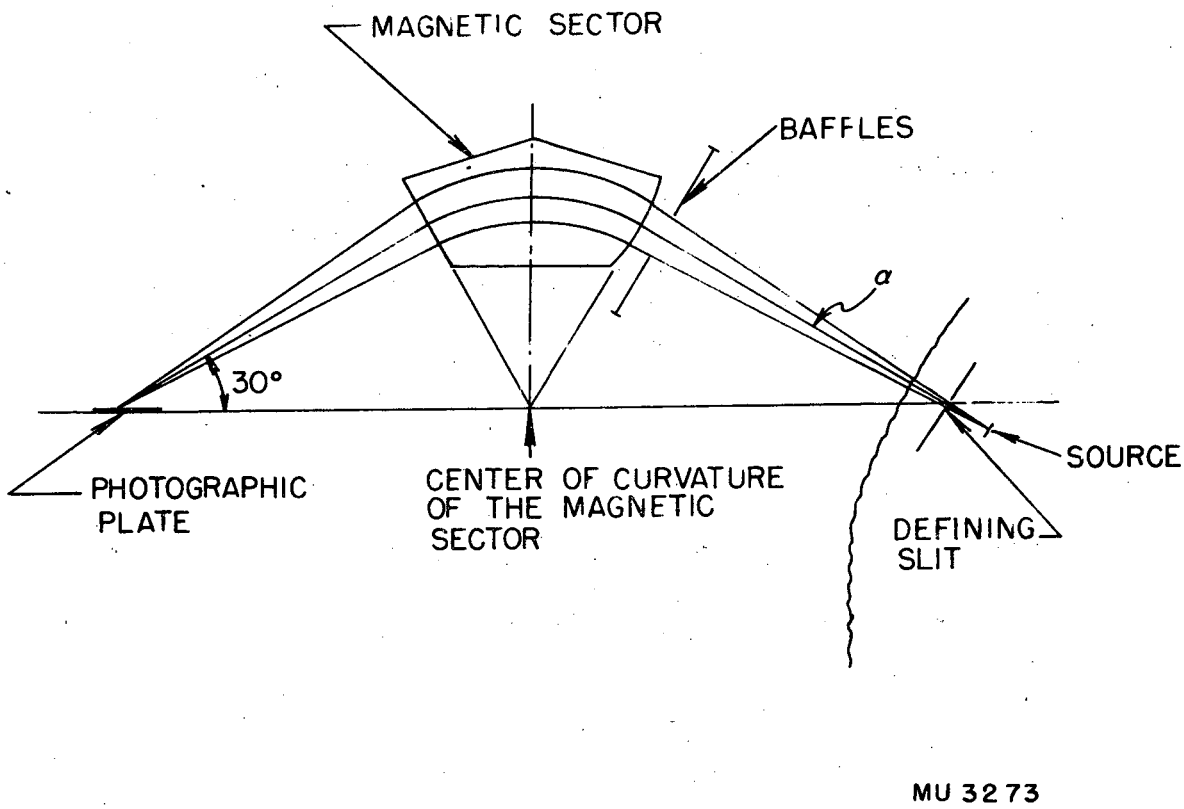

Fig. 1. Schematic diagram of optics of spectrograph.

fluctuations are monitored constantly with a Speedomax recorder. The vacuum in the spectrograph is maintained by two 260 liter/sec oil diffusion pumps each backed by Duo Seal mechanical pump. The source and detector portions of the spectrograph, which may be sealed off from the main vacuum tank for the purpose of changing samples or photographic plates, have separate mechanical roughing pumps. The photographic plate holder or receiver is shown in Fig. 2. It holds two 9 in. x 2 in. photographic plates which may be exposed successively without letting the receiver down to air. It has a light-tight camera door which may be operated outside the spectrograph after the receiver has been evacuated.

The source and slit holder with two 0.018 in. slits and a 1 in. spacer is shown in Fig. 3. Various combinations of 1/8 in., 0.018 in., 0.010 in., and 0.005 in. slits separated by 1/4 in. or 1 in. spacers in conjunction with a baffle system close to the magnet gap permitted variations in solid angle from 10^{-4} to 10^{-7} of 4π.

1. Sample preparation. -- In order to take advantage of the inherent high resolution of an alpha particle spectrograph, it is imperative that the sources be extremely thin. Poor samples with respect to self-absorption are manifested by a tail on the low energy side of the distribution curve, whereas the form for a thin preparation approaches a symmetrical peak. Among the natural radioactivities good samples have been prepared when applicable by collecting the active deposits from the emanation decay series.

In general, poor samples are obtained when prepared from solutions by simple evaporation of the solvents. Even when the

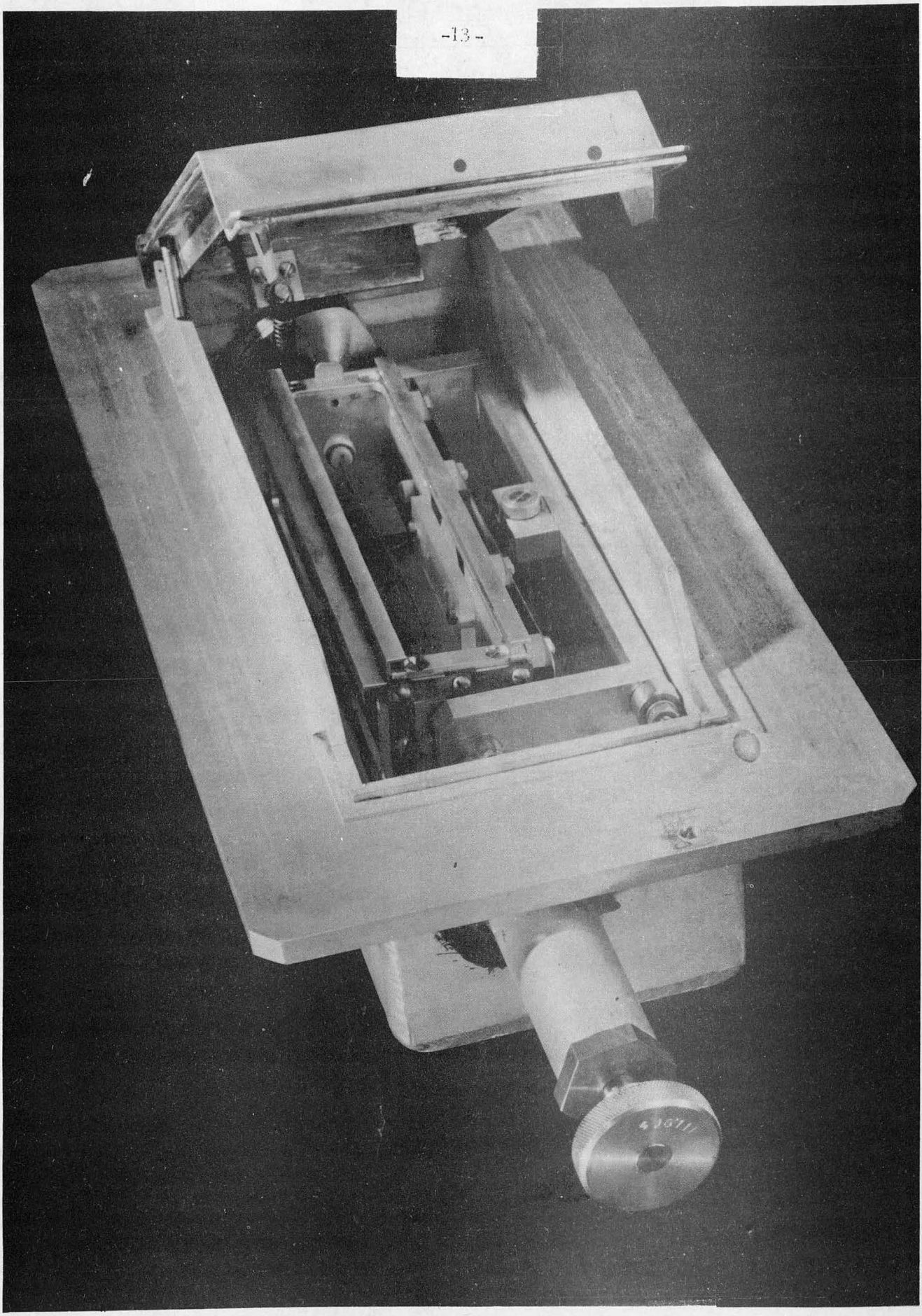

Fig. 2. Photograph of receiver.

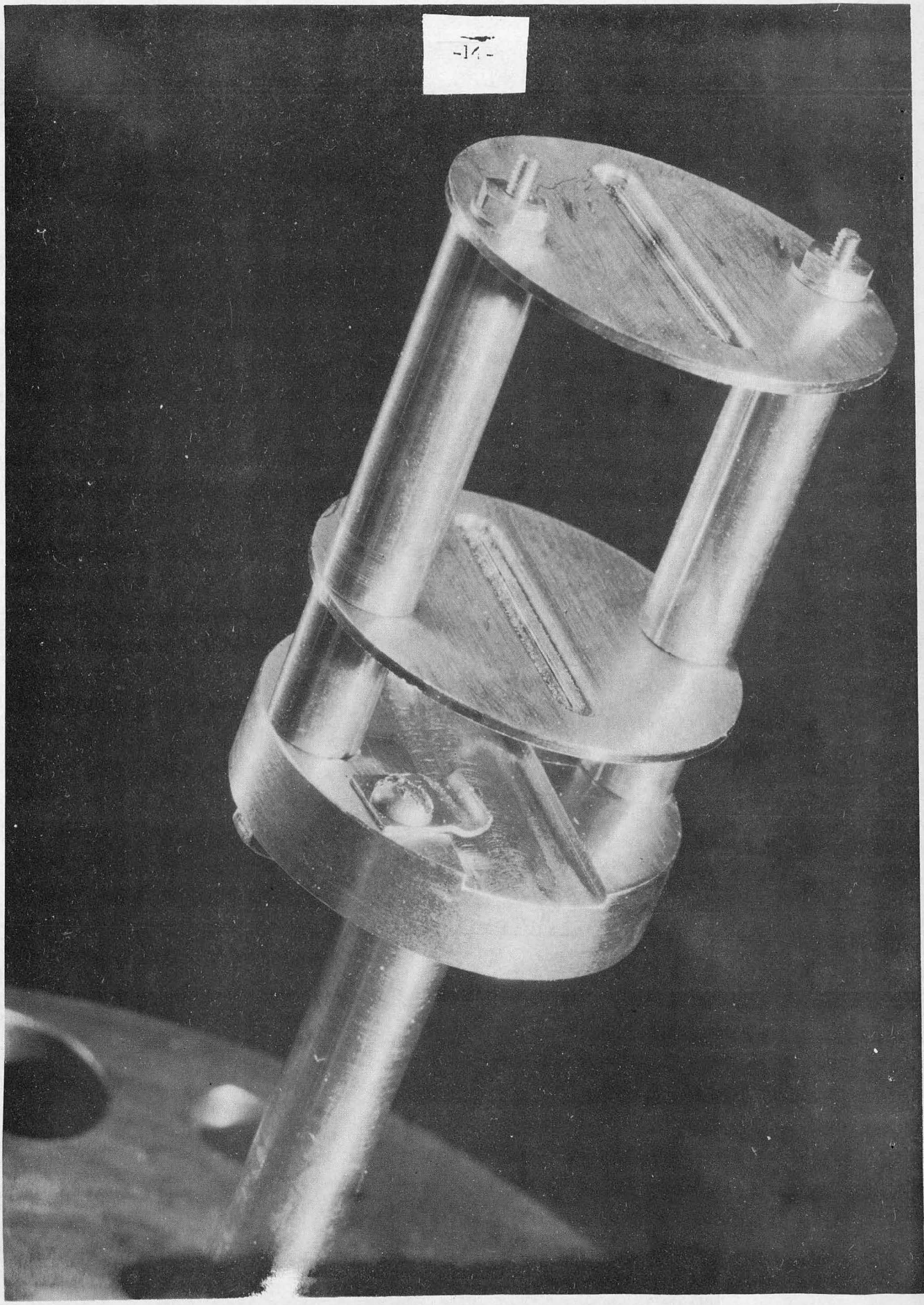

Fig. 3. Photograph of source.

weight of the sample corresponds only to the order of a few micrograms per square centimeter on the plate, the formation of microcrystals with high surface density effectively produces a thick sample. Impurities in the solution often produce the same results. In certain cases electrodeposition may be employed to advantage but in general this method requires careful control of conditions which may be different for each substance. It is difficult to obtain satisfactory plates, particularly for electropositive elements such as the actinide elements in which reduction to the metal is not possible. The most generally acceptable method employed in this laboratory for a wide variety of substances consists of vacuum sublimation. In the presently considered instances, solutions of radioactive chlorides, nitrates, or citrates are evaporated to dryness on a tungsten filament. Upon raising the temperature to white heat for a few seconds by passing current through the filament, the sample is vaporized onto a 2 mil platinum plate masked by another plate having a rectangular slit 1 in. x 1/8 in. which defines the sample shape. The mask and collecting plate are placed about 1/8 in. above the filament. The whole system is maintained at a few microns pressure to reduce formation of tungsten oxide which, when formed, appears as a dark film on the collecting plate. Under optimum conditions, the vaporized sample can contain up to 50 $\mu g/cm^2$ of active atoms and still give adequate resolution with very little low energy tailing. For high resolution the limit is generally under 10 $\mu g/cm^2$. By shaping the tungsten filament into a trough, sublimation yields of 50 percent are generally obtained on the collecting plate.

2. Sample exposure and track detection. --By reference to Fig. 1, it may be seen that the alpha particle beam is defined by the slit and baffle system as indicated. With a uniform magnetic field, the width (s) of the image on the photographic plate of a homogeneous beam of alpha particles is:

$$S = 2(\Delta S + r_o \alpha^2 + \ldots\ldots), \qquad (1)$$

where ΔS is the defining slit width, r_o the radius of the normal trajectory, and α the half angle of emergence of the beam from the magnetic field. This formula applies to a 60° sector magnet with plane surfaces for the pole pieces and the factor "2" arises because the photographic plate is placed at an angle of 30° with the trajectory of the alpha particles rather than normal to it.

By proper shaping of the source side of the magnet, the second term of equation (1) can be made negligible with respect to the slit width, and this feature is indicated on the diagram by the rounded surface of the magnet. Since the magnetic field is not uniform throughout, but falls off near the edges of the magnet, baffles are used as a means of confining the beam to the center. If a single slit opening of 0.018 in. and a 3 in. opening between the baffles are used, the alpha beam half-width on the photographic plate for a 6 Mev alpha particle is 2 mm which corresponds to 8 kev.

Sample strengths are in general selected to permit exposures of one to two days. However, exposures as short as 6 minutes and as long as 16 days have been employed. The lengthy exposures were required to examine the alpha groups

present in low abundance in complex spectra. A single field of view in the microscope encompasses 1/4 x 1/4 mm of the photographic plate, and a scan of one field width across the entire height of the plate (2 inches) gave as background two alpha tracks for each day's exposure. This background, however, would vary considerably depending on the amount of alpha contamination in the instrument. On the low energy side of an alpha peak, the apparent background could be considerably higher than stated above, presumably because of the low energy tailing due to absorption in the sample or scattering in the spectrograph.

As already mentioned, the alpha particles are determined photographically. The detecting plates used were 9 in. x 2 in. Eastman NTA plates with 25 or 30 micron thick emulsions and these were examined under a 450 power microscope with bright field illumination. The track length of a 6 Mev alpha particle is approximately 25 microns, and because of the position of the plate as shown in Fig. 1, the track makes an angle of 30° with the plane of the emulsion. Because of the small angle of acceptance parallel to the magnetic field permitted by the gap between the pole pieces, the tracks should be nearly parallel and only these are recorded. A photograph of one field of view is shown in Fig. 4 in which there are seen six acceptable alpha tracks and two tracks which are rejected.

In the measurements made in the early part of this work, the particular microscope stage could not hold a 9 in. plate so each

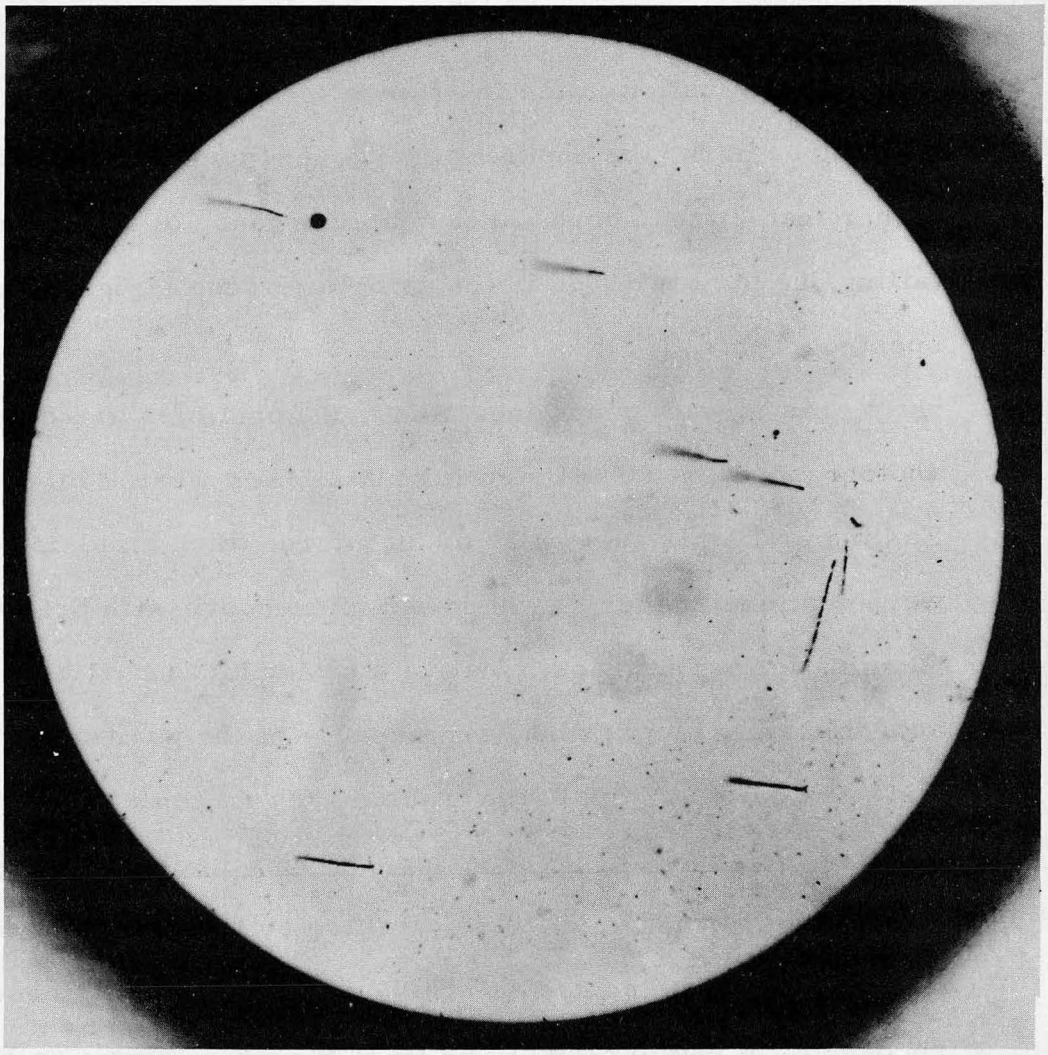

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Fig. 4. Photograph of one microscope field of view

plate was cut into three parts. Before it was sectioned, an axial line was ruled with a razor blade along the center of the plate parallel to the long dimension. Cross lines perpendicular to the axial line were also ruled in each of the proposed sections. The distances between the intersections of the lines were measured, and after sectioning these served as indices to relate distances on the three sections. In the later portions of this work, a stage capable of holding a 9 in. plate was obtained. Again an axial line was ruled along the center of the plate parallel to the long dimension. One cross line was drawn perpendicular to the axial line and the resulting intersection served as a reference point so that the plate could be removed and then replaced in exactly the same position.

In counting the tracks the microscope stage was moved perpendicular to the axial line giving a scan 1/4 mm wide across the width of the plate. The stage was then moved one field of view parallel to the axial line and another scan made. The count from each scan was plotted on a count versus distance graph as shown in Fig. 7.

3. Dispersion. -- The energy dispersion on the plate for a normal trajectory, r_o, and energy, E_o, is

$$\text{dispersion} = E_o/2r_o. \qquad (2)$$

The relation of the magnetic field to E_o and r_o is given by equation (3) in which B is in gauss, r_o in centimeters and E_o in electron volts.

$$B = \frac{144}{r_o} E_o^{1/2} \qquad (3)$$

Since the position of the normal trajectory cannot be determined

precisely, it is not possible to determine the value of B necessary to focus a particular alpha particle at that point and E_o is best eliminated between equations (2) and (3). The resulting equation (4) for the dispersion is then:

$$\text{dispersion} = r_o B^2/2 \times (144)^2 . \qquad (4)$$

In addition, r_o cannot be expected to be precisely equal to the nominal radius of curvature of the magnet (75 cm) because of lack of precision in construction and alignment of source and detector. If the magnetic field is known accurately, however, an effective radius can be determined by measuring distances between alpha groups of known energy. In other words, the dispersion is obtained experimentally for the particular energy range of interest. For this purpose, measurements were made of the separation between the two alpha groups of Ra^{226}, the separation between Em^{222} and Po^{218} and the separation between Po^{210} and Em^{222} as shown in Fig. 5a. The energies taken for Em^{222} and Po^{218} alpha particles were those given by Briggs (5.486 and 5.998 Mev)[2] and for the two Ra^{226} groups the separation given by Rosenblum[3] as 188 kev. The energy taken for Po^{210} was 5.298 Mev.[4,5] Within the limits of experimental error (about 2 percent of the energy differences) the radius so determined was constant and indicated the nominal 75 cm radius must be increased by 5.9 percent; that is, energy differences calculated by the use of equation (4) using 75 cm radius were low by this amount.

In the beginning stages of this study, the magnetic field was measured as a function of the magnet current. The magnetic field

was determined on subsequent runs by accurately measuring the magnet current and reading the field from the field-current relation. In the later stages of this research, however, a Varian Associates proton fluxmeter, Model F6, was used to measure the field before every exposure.

In order to measure the separation between peaks, the high energy sides of the peaks were extrapolated to the background. The separation used was the difference between the background intersections. Since the half-width of the alpha peaks increases appreciably toward the ends of the photographic plate and since the high energy side is particularly distorted at the plate ends, the above method of determining separations was subject to some error. In order to check the effect of the distortion on the energy measurement, six sets of Cm^{242} peaks were spaced at intervals along a single photographic plate. Each Cm^{242} set consisted of two alpha groups separated by a constant energy. With the use of the above method of determining peak separations, very erratic differences in energy separation were observed for the various sets.

With the thought that this apparent erratic energy deviation might be affected to some extent by changes in the dispersion along the plate, the dispersion was calculated as a function of position on the photographic plate and the calculated variations were small compared to the experimental errors involved.

It was found that the separation determined by the difference between the tips of the peaks of each set, or the intersection of extrapolations of the high and low energy sides of the peak gave constant energy differences for the various sets of peaks within

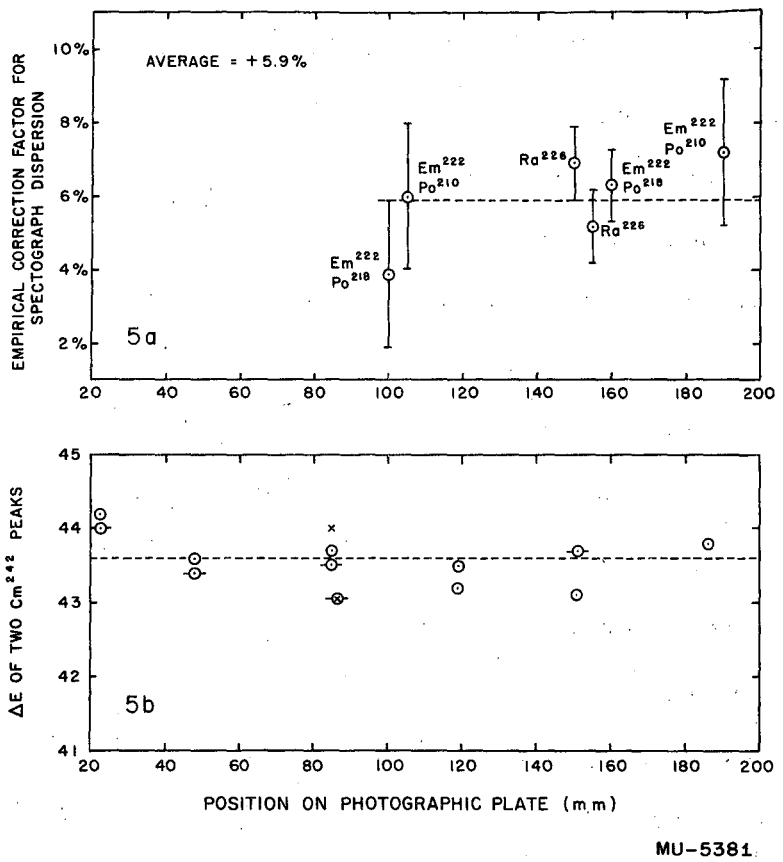

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Fig. 5. a) Energy calibration.
b) Uniformity of energy calibration.

experimental error, as shown in Fig. 5b. Therefore, this latter method of determining energy separations has been used for all of the later work.

In most of the early cases studied, the alpha peaks were in the center of the photographic plate and no appreciable error would be introduced by determining energy differences from extrapolations of the high energy edge. In the determination of the alpha particle energy of Am^{241} against Po^{210} and Em^{222}, the Po^{210} peak in the former case and the Em^{222} and Am^{241} peaks in the latter case were very close to the end of the plate and the high energy tailing is visible in Fig. 18 and 19.

Small changes in the alignment of the source and detector would shift the high energy tailing to various portions of the photographic plate; for example, Fig. 7 and 8b are graphs of spectra taken while the instrument alignment was rather poor. Fig. 8a, 8c and 8d are graphs of spectra taken after a better alignment had been made.

B. The Alpha-Gamma Coincidence Counter

A block diagram of the electronics circuit involved in the alpha-gamma coincidence counter is shown in Fig. 6. After amplification and shaping, the pulses from the gamma detector are fed into a one-channel Schmidt type differential discriminator. This discriminator contains variable lower and upper biases which eliminate all pulses except those in a definite pulse height region. In normal operation the voltage separation between the lower and upper biases remains constant while their absolute voltage is being

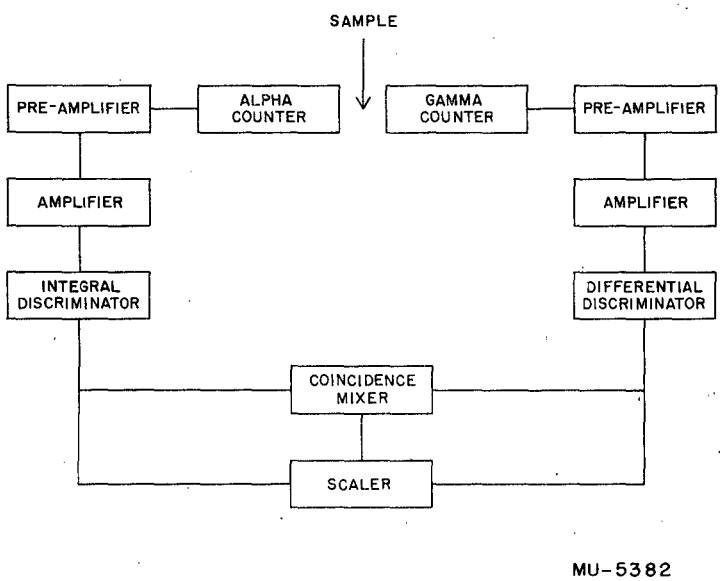


Fig. 6. Block diagram of the alpha-gamma coincidence counter circuits.

changed. The output of the differential discriminator is fed into the coincidence mixer and also into a scaler. The pulses from the alpha detector are fed into a Schmidt type discriminator after amplification and shaping. This discriminator consists of a single variable bias to eliminate pulses of very low energy. The output of this discriminator feeds into the coincidence mixer and into the scaler. The output of the coincidence mixer feeds into the scaler also.

1. The gamma detector. --The gamma detector consists of a Tl activated NaI crystal mounted Borkowski fashion onto an RCA 5819 Photomultiplier tube. The crystal is a solid cylinder 1 in. in diameter and about 5/8 in. thick. One of the plane faces was whittled to fit onto the end of the photomultiplier tube. The crystal was attached to the tube with white vaseline and then placed in a cylindrical copper can 2 1/4 in. in diameter and about 4 in. long. One end of the can was closed with a 0.002 in. copper window. Before pushing the crystal and photomultiplier tube into the can it had been partially filled with MgO powder so that the crystal would be entirely surrounded by the powder except for the face attached to the tube. The MgO cuts down the light loss from the crystal and thus permits higher pulses from the photomultiplier tube. The can was then sealed with vacuum putty and wrapped with black scotch tape.

2. The alpha detector. -- The alpha detector consisted simply of a ZnS screen sprayed onto a lucite light pipe which was mounted in front of an RCA 5819 photomultiplier tube.

3. Operating technique. -- The alpha-gamma coincidence counter could be used as a one-channel gamma analyzer, or coincidences could be determined between gamma rays in a given channel and the entire alpha spectrum.

When used simply as a gamma analyzer, the gamma counting rate is determined for various voltage settings of the biases of the differential discriminator. The counting rates are then plotted as a function of the discriminator reading as shown in Fig. 9. In order to evaluate the energies of the gamma peaks, the spectra are compared with the Am^{241} 60 kev gamma and the U^{235} 184 kev gamma. The energy dispersion is obtained by extrapolating the sides of the peaks to their intersections, determining the separation between the 60 kev and 184 kev peak intersections in discriminator volts, and dividing the energy separation (184 - 60 = 124 kev) by the separation in volts. The energies of any other gammas are then determined by (1) extrapolating the sides to their intersection; (2) finding the separation in volts between their intersection and the intersection of either the Am^{241} 60 kev gamma ray or the U^{235} 184 kev gamma ray, (3) multiplying the voltage separation by the dispersion and adding to or subtracting from the energy of the standard as is applicable.

When used as an alpha-gamma coincidence counter, the operating technique was much the same as above, with the following exceptions. The alpha active sample on a platinum or aluminum plate usually would be placed on a cardboard backing about 1/8 in. from the ZnS screen, the whole assembly being light-tight. The gamma detector would then be placed directly behind and touching

the cardboard backing.

At each discriminator setting, the alpha, gamma, and coincidence counting rates were determined. This was necessary to determine the chance coincidence rate. This chance coincidence rate obeys Poisson's Law, but a rather simple approximation is:

$$\text{Chance Coincidences} = (\text{Resolving time of Instrument})\ (\text{Gamma Counting Rate})\ (\text{Alpha Counting Rate})$$

The resolving time of the instrument was determined to be 2.5 microseconds by counting chance coincidences resulting from independent alpha and gamma sources.

4. Geometry. -- The geometry of the unit was determined for each gamma or coincidence experiment with the Am^{241} 60 kev gamma ray. The abundance of this gamma ray was taken as 0.40 gammas per alpha,[6] each gamma being in coincidence with an alpha particle.

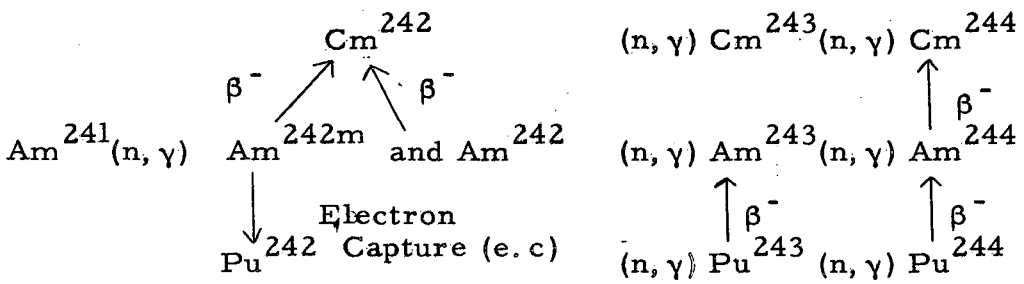
5. Linearity and stability. -- With the 60 kev gamma ray of Am^{241}, the 184 kev gamma of U^{235} and the L x-rays of the heavy elements used as standards, it was found that the pulse height from the gamma detector was linear with energy in the region studied.

Over a period of a week, the peak position of a gamma ray would drift by about two volts. But even over short periods of time shifts of 1 or 2 volts took place, so for very accurate energy measurements more than one calibration was necessary.

C. Complex Alpha Spectra of $Cm^{242, 243, 244}$

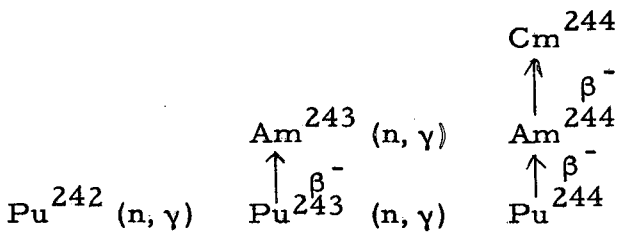
The isotopes Cm^{242}, Cm^{243}, and Cm^{244} can be produced in quantities adequate for the spectrograph by pile bombardment of Am^{241}.

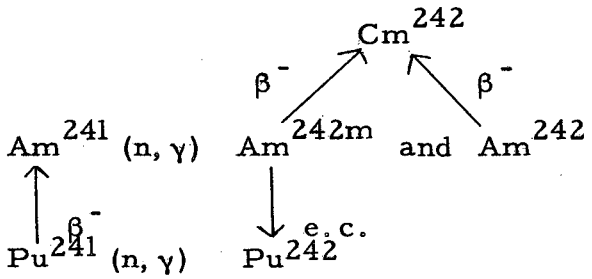
The reactions would be:



In this case, most of the alpha activity would be due to Cm^{242}.

The isotopes Cm^{242} and Cm^{244} were prepared without Cm^{243} by bombardment of plutonium enriched in Pu^{242}. The reactions would be:





In the short period of time which the plutonium spent in the pile, comparable quantities of Cm^{242} and Cm^{244} would be formed with very little Cm^{243}.

The curium fractions were purified chemically from all heavy element alpha emitters except traces of Am^{241} and some Pu^{238} which grows in from the decay of Cm^{242}. The Am^{241} and Pu^{238} alpha spectra have been studied separately, as will be mentioned in a later section, so their contribution may be subtracted from the curium alpha spectra.

The alpha spectrum of Cm^{242} has been mentioned previously[7] as consisting of two alpha groups; the most energetic having an abundance of 73 percent and the other having an abundance of 27 ± 2 percent. These groups were separated by 46.5 ± 1.0 kev in decay energy.

In the previous measurement cited,[7] the energy separation between the two alpha groups of Cm^{242} had been determined by extrapolating the high energy sides of the peaks to the background. Since that time, the spectrograph has been disassembled, reassembled and realigned, and the proton resonance fluxmeter has been installed.

After the above improvements were made, a sample of Cm^{242} was run in the spectrograph at six different magnetic field settings on the same photographic plate, thus giving six sets of Cm^{242} peaks. In Fig. 5b, the energy separation between the two alpha groups of each set is plotted against the position on the photographic plate of the midpoint of the peaks.

It is seen that within the limits of determining the peak position, about one field of view in the microscope, or 1 kev, the energy separation is constant over most of the photographic plate.

The energy separation between these alpha groups of Cm^{242} is taken as the average of the 13 values shown in Fig. 5b, 43.6 ± 0.6 kev, with the limits of error in determining the peak position including all values. The errors in calibration as seen on Fig. 5a are all less than 2 percent, so 2 percent will be taken as the limit of calibration error, thus giving a particle energy separation of 43.6 ± 1.4 kev.

With the correction for the difference in recoil energy associated with the two alpha groups, the alpha decay separation becomes 44.3 ± 1.4 kev. According to the nomenclature mentioned in an earlier paper,[7] we will designate the alpha group leading to the daughter ground state as α_o of Cm^{242} and the alpha group leading to the state 44.3 kev above the daughter ground state as α_{44} of Cm^{242}. Further relative abundance measurements have been made on this spectrum and show that the abundance of α_{44} of Cm^{242} is 26.3 ± 0.5 percent. This value is the average of our six best measurements and the limits of error include all the measurements.

In the first report made on this alpha spectrum,[7] the energy of Cm^{242} α_o was reported as 6.110 ± 0.003 Mev from comparison with the alpha group of Po^{218}. The particle energy of the latter nuclide has been measured by G. H. Briggs[2] as 5.998 Mev.

In order to verify this determination, the energy of α_o of Cm^{242} was compared with the main group of Am^{241}. The value used for the energy of the Am^{241} main group was 5.476 Mev from data presented in a following section. Based on the Am^{241} comparison, the energy of Cm^{242} α_o was 6.112 ± 0.010 Mev. With the limits of

error as given, the close agreement was fortuitous and the value selected was that of the more accurate measurement, 6.110 ± 0.002 Mev. Since this group is probably that of the ground state transition, the decay energy of Cm^{242} is accordingly 6.211 Mev. Our results are in moderate agreement with those of other workers who, using ion chambers, have reported particle energies of 6.118[8] and 6.08[9] Mev for Cm^{242}.

Since our first publication,[7] improved techniques in sample preparation have resulted in less tailing on the low energy side of intense alpha groups, thus permitting a detailed investigation of low intensity alpha groups of Cm^{242}, Cm^{243} and Cm^{244}.

A sample of curium was obtained which had been prepared by bombardment in a pile of a mixture of $Pu^{238, 239, 240, 241, 242}$ enriched in Pu^{242}.

A sample of curium suitable for the spectrograph was prepared by sublimation and the alpha activity of about 10^6 dis/min covered an area 1 in. x 1/8 in. The sample, marked by a 1 in. x 0.018 in. stainless steel slit, was run in the spectrograph for 42 hours and its alpha spectrum is shown in Fig. 7. The two highest energy peaks are due to Cm^{242}, since their energy separation, relative abundance, and position on the photographic plate all correspond to the known Cm^{242} alpha spectrum.

The particle energies of all alpha groups on Fig. 7, 8a, 8b, 8c, and 8d were determined using 6.110 Mev as the energy of α_0 of Cm^{242}, or 6.066 Mev as the energy of α_{44}. Table 1 is a list of the energies of all the alpha groups observed in Fig. 7 and

Table 1

Energies and Abundances of $Cm^{242, 244}$ Alpha Spectra

Energy of alpha group in Fig. 7 (Mev)	Abundance of alpha group with respect to the total Cm^{242} alpha activity (%)
5.73 → 5.74	< 1.4[a]
5.755	11
5.775 → 5.785	< 2[a]
5.797	35
5.98 → 5.99	<1.4[a]
6.00 → 6.01	<1.4[a]
6.025 → 6.035	<2[a]
6.066 } 6.110 }	100

[a]Determined by comparing peak heights.

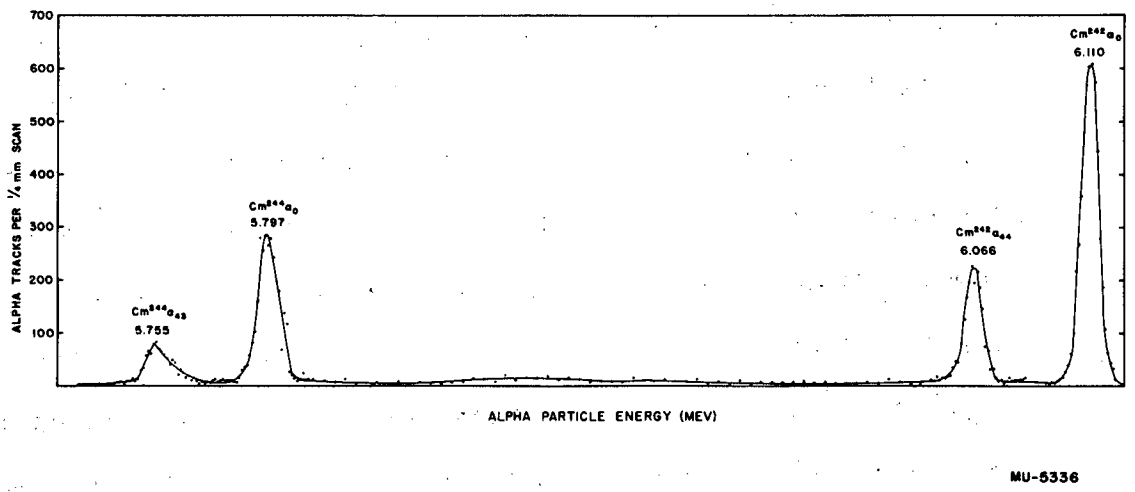


Fig. 7. $Cm^{242,244}$ alpha spectra.

and their abundance with respect to Cm^{242}. The maximum abundances of any unresolved peaks at energies corresponding to observed alpha groups on subsequent plates are also included.

Another quantity of curium was obtained which had been prepared by bombarding Am^{241} in a pile. A sample was prepared for the spectrograph by sublimation and the alpha spectra are shown in Fig. 8a. The alpha activity of the sample was 1.5×10^9 dis/min and the length of the spectrograph run was 14 hours 19 minutes. A 1 in. x 0.018 in. stainless steel slit was used to mask the sample in the spectrograph. Table 2 shows the energies and abundances of the various alpha groups observed in Fig. 8a.

A third quantity of curium was obtained which had been prepared by bombarding in a pile the curium stock from which the spectrograph sample whose spectra are shown in Fig. 8a was made. A spectrograph sample of 6×10^8 dis/min was prepared by sublimation and the alpha spectra are shown in Fig. 8b. The length of the exposure was 16 hours 18 minutes, and the sample was masked in the spectrograph by a 1 in. x 0.018 in. stainless steel slit. Table 3 shows the energies and abundances of the various alpha groups observed in Fig. 8b.

The sample whose alpha spectra are shown in Fig. 8b was rerun in the spectrograph for 61 hours 15 minutes starting the next day. These spectra are not shown. Table 4 shows the energies and abundances of the various alpha groups observed in this exposure and the maximum abundances of any groups which appear in other figures.

Table 2

Energies and Abundances of $Cm^{242, 243, 244}$ Alpha Spectra Seen in Fig. 8a

Energy of the Alpha Groups in Fig. 8a (Mev)	Abundance of the Alpha Groups with Respect to the Total Cm^{242} Alpha Activity (%)
5.734	0.065
5.757	0.091
5.778	0.43
5.800	0.31
5.965	0.033
5.986	0.029
6.006	0.005
6.029	0.006
6.066 } 6.110 }	100

Energies and Abundances of $Cm^{242, 243, 244}$ Alpha Spectra Seen in Fig. 8b

Energy of the Alpha Groups in Fig. 8b (Mev)	Abundance of the Alpha Groups with Respect to the Total Cm^{242} Alpha Activity (%)
5.728	0.020
5.751	0.053
5.774	0.12
5.795	0.17
5.964	0.035
5.985	0.011
6.006	0.007
6.030	0.004
6.066 } 6.110 }	100

Table 4

Energies and Abundances of $Cm^{242, 243, 244}$ Alpha Spectra Not Shown

Energy of the alpha groups in spectra not shown (Mev)	Abundance of the alpha groups with respect to the total Cm^{242} alpha activity (%)
5.729	0.021
5.755	0.067
5.776_5	0.11
5.797	0.18
5.963	0.035
5.983	0.0064
6.00 → 6.01	<0.004[a]
6.025 → 6.035	<0.003[a]
6.066, 6.110	100

[a]Determined by comparing peak heights.

After a period of 90 days, the sample whose spectra are shown in Fig. 8b was exposed again. The length of the exposure was 63 hours 42 minutes and its alpha spectrum is shown in Fig. 8c. The sample was masked in the spectrograph by a 1 in. by 0.005 in. stainless steel slit. Table 5 shows the energies and abundances of the various alpha groups observed in Fig. 8c and the maximum abundance of any groups which appear in other figures.

A fourth quantity of curium was obtained which had been prepared from an Am^{241} pile bombardment. A sublimed sample of 3×10^8 dis/min was prepared and exposed in the spectrograph for 44 hours. Its spectra are shown in Fig. 8d. Table 6 shows the energies and abundances of the various alpha groups observed in Fig. 8d and the maximum abundance of any groups which appear in other figures.

1. Principal groups of Cm^{244}. -- From the bombardment conditions under which the curium whose alpha spectra are shown in Fig. 7 was made, only Cm^{242} and Cm^{244} would be prepared. Hence the alpha groups at 5.797 and 5.755 Mev must belong to Cm^{242} or Cm^{244}. These alpha groups were also seen on Figs. 8a, 8b, 8c and 8d. The average particle energies were 5.798 and 5.755 Mev. Table 7 shows the abundances of these groups for the various exposures. Since the abundance of the 5.798 Mev group relative to the Cm^{242} activity varies by over two orders of magnitude in the various exposures shown in Table 7, this alpha group obviously belongs to Cm^{244} and will be designated Cm^{244} α_0. This result is in excellent agreement with the previous ion chamber measurement of 5.79 Mev made by Thompson, Ghiorso and Reynolds.[10]

Table 5

Energies and Abundances of $Cm^{242, 243, 244}$ Alpha Spectra Shown in Fig. 8c

Energy of the alpha groups in Fig. 8c (Mev)	Abundance of the alpha groups with respect to the total Cm^{242} alpha activity (%)
5.735	0.028
5.758	0.084
5.779	0.20
5.800	0.26
5.966	0.036
5.986	0.017
6.00 → 6.01	<0.005[a]
6.025 → 6.035	<0.007[a]
6.066, 6.110	100

[a]Determined by comparing peak heights.

Table 6

Energies and Abundances of $Cm^{242, 243, 244}$ Alpha Spectra Shown in Fig. 8d

Energy of the alpha groups in Fig. 8d (Mev)	Abundance of the alpha groups with respect to the total Cm^{242} alpha activity (%)
5.725 → 5.735	<0.007[a]
5.756	0.0205
5.776	0.035
5.797	0.050
5.964	0.037
5.98 → 5.99	<0.006[a]
6.00 → 6.01	<0.009[a]
6.025 → 6.035	<0.01[a]
6.066, 6.110	100

[a]Determined by comparing peak heights.

Table 7
Abundances of the Curium 5.798 and 5.753 Mev Groups with Respect to Other Groups

	Fig. 7	Fig. 8a	Fig. 8b	Fig. 8c	Fig. 8d	Spectra not shown
Abundance (%) of the 5.798 Mev group relative to the sum of the Cm^{242} groups	35	0.31	0.17	0.26	0.05	0.18
Abundance (%) of the 5.755 Mev group relative to the sum of the intensities of the 5.798 Mev group and the 5.755 Mev group	24.0	22.7	23.7	24.9	28.5	27.6

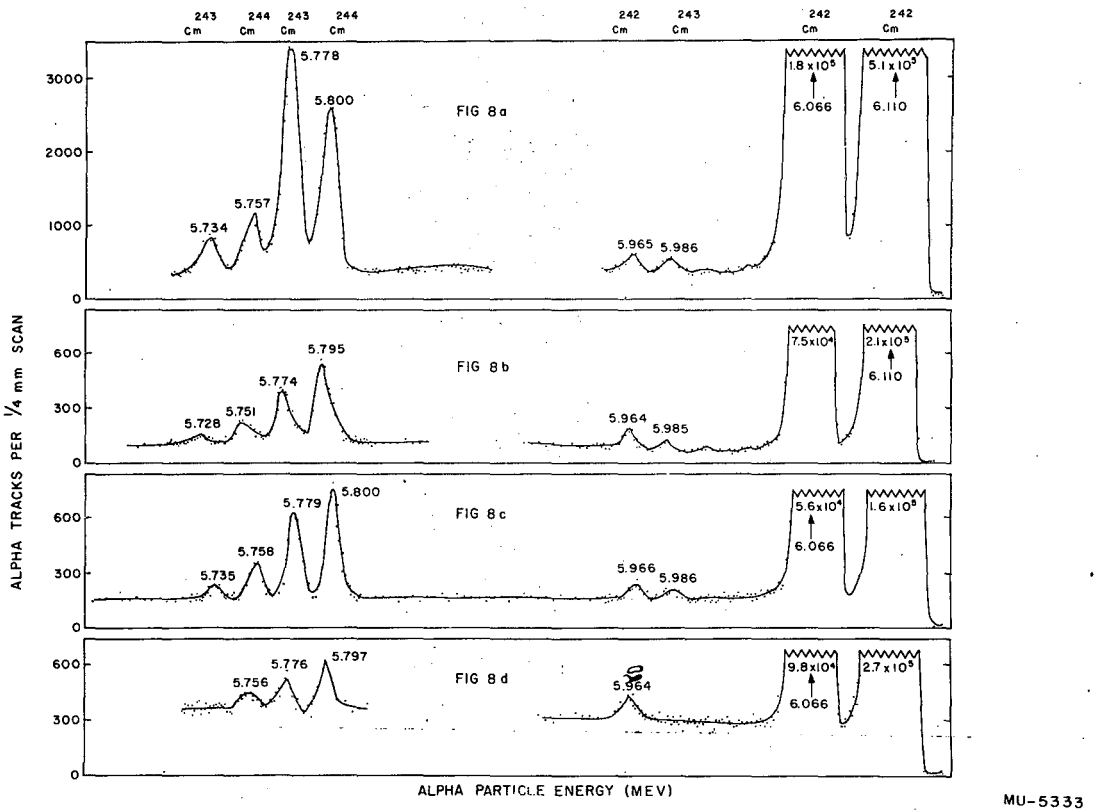


Fig. 8. $Cm^{242, 243, 244}$ alpha spectra.

Since the abundance of the 5.755 Mev group relative to the sum of the abundance of the 5.798 and 5.755 Mev groups is nearly constant for the various exposures (25 ± 3%), the 5.755 Mev group also belongs to Cm^{244}. The average separation between Cm^{244} α_0 and the 5.755 Mev group is 42.5 ± 1.7 kev with limits of error including all values. Upon the addition of a 2 percent possible calibration error, we obtain 42.5 ± 2.5 kev. Adding the difference in recoil energy between the two groups, we find the alpha decay separation to be 42.5 + 0.7 kev = 43.2 kev. Therefore the 5.755 Mev group is designated α_{43} of Cm^{244}.

2. Low energy alpha groups of Cm^{242}. -- In the early portion of this research[7] it was found that at energies lower than the two principal groups (6.110 and 6.066 Mev) there was no alpha group in abundance greater than about 0.1 percent. With the improvements made in experimental technique, a group has appeared at 5.964 Mev and is assigned to Cm^{242}. This group appears in all of the spectra of Fig. 8. The experimental proof that it arises from Cm^{242} decay lies in the invariance of its abundance relative to the principal groups of Cm^{242}. The data are summarized in Table 8 where this fixed ratio is apparent, whereas the ratio of this group at 5.964 Mev to groups ascribed to Cm^{243} and Cm^{244} undergoes considerable variation.

Two other possible groups have shown up (Fig. 8a and 8b) at 6.006 and 6.029 Mev and appear to remain invariant to the abundance of the principal groups of Cm^{242} rather than Cm^{243} or Cm^{244}. It cannot be definitely concluded, however, that they

Table 8

Abundances of the Curium 5.964 Mev Group
With Respect to Other Groups

	Fig. 7	Fig. 8a	Fig. 8b	Fig. 8c	Fig. 8d	Spectra not Shown
Abundance (%) of the 5.964 Mev group relative to Cm^{242}	<1.4	0.033	0.035	0.036	0.037_5	0.035
Abundance (%) of the 5.964 Mev group relative to Cm^{244}	<3	8.2	16	11	53	14
Abundance (%) of the 5.964 Mev group relative to the 5.777 Mev group		7.7	29.5	18	107	32

are emitted at those energies by the Cm^{242} nucleus since their low intensity does not permit ruling out instrumental effects.

3. Alpha groups of Cm^{243}. -- It remains now to assign groups at 5.985, 5.777 and 5.732 Mev which appear in Fig. 8a, 8b, 8c and 8d. The pertinent numerical data of Fig. 7, 8a, 8b, 8c and 8d are summarized in Table 9 in the form of ratios of abundances for the various groups. The group at 5.777 Mev is the most prominent and is used as the reference. Within the limits of error in the measurements, the ratio of the three groups among themselves are constant.

On the contrary, the ratio in abundance of the 5.777 Mev group to the Cm^{242} activity varies by almost a factor of 10 and varies by an even larger factor with respect to the Cm^{244} activity. On this basis alone it is probable that all three groups belong to Cm^{243}. The relative abundances of the 5.985, 5.777 and 5.732 Mev groups are 6, 81 and 13 percent, respectively.

Prior to this study, ion chamber measurements by Thompson, Ghiorso and Seaborg[11] gave energies of 5.79 Mev (85 percent) and 5.89 Mev (15 percent). Although the 5.79 Mev value is in good agreement with our work, the 5.89 Mev energy disagrees by nearly 90 kev with our value.

This anomaly might be explained if one assumes that the conversion electrons in coincidence with the Cm^{243} alpha groups around 5.79 Mev are losing a portion of their energy in the ion chamber in which the alpha particle energies are being measured. This could cause a high energy tail of considerable proportions since the coincident radiations have energies up to 278 kev as will

Table 9

Abundances of the 5.985, 5.777 and 5.732 Groups

	Fig. 7	Fig. 8a	Fig. 8b	Fig. 8c	Fig. 8d	Spectra not shown
Ratios of the relative abundance of the 5.985, 5.777 and 5.732 Mev groups		0.068:1:0.15	0.11:1:0.17	0.083:1:0.14	<0.17:1:<0.21	0.049:1:0.19
Abundance (%) of the 5.777 Mev group relative to Cm^{242}	<1.7	0.43	0.12	0.20	0.035	0.11
Abundance (%) of the 5.777 Mev group relative to Cm^{244}	<3.6	106	54	59	49	45

be shown in a later section.

From a knowledge of the relative alpha activities and masses of the various curium isotopes, the half-life of Cm^{243} and Cm^{244} can be determined from the half-life of Cm^{242}. A representative equation for Cm^{244} is:

$$\begin{pmatrix}\text{Half-life}\\ Cm^{244}\end{pmatrix} = \begin{pmatrix}\text{Half-life}\\ Cm^{242}\end{pmatrix}\begin{pmatrix}\text{Ratio of alpha}\\ \text{activity of } Cm^{242}\\ \text{to } Cm^{244}\end{pmatrix}\begin{pmatrix}\text{Ratio of atoms of}\\ Cm^{244} \text{ to } Cm^{242}\end{pmatrix}$$

The data on the atomic ratios were obtained by F. L. Reynolds of this laboratory with a mass spectrometer. The atomic and alpha particle abundance ratios were determined for the curium whose alpha spectra are shown in Fig. 8d. The respective half-lives for Cm^{243} and Cm^{244} are 35 and 19 years. The latter number is in excellent agreement with the half-life of 19 years found by Thompson, Hulet and Ghiorso[12] by decay measurements.

D. Gamma Spectra of Cm^{242} and Cm^{243}

The gamma spectra of the samples whose alpha spectra are shown in Fig. 8a and 8d were studied with the one-channel gamma analyzer described in an earlier section. The gamma spectra of these samples are shown in Fig. 9 and 10.

In addition, a sample of Cm^{242} of very high isotopic purity was obtained from G. H. Higgins and E. K. Hulet. This curium gamma spectrum was measured with the analyzer described previously. This latter spectrum was also studied with a 40-channel gamma analyzer and one of the consequent spectra is shown in Fig. 11.

1. Assignment of gamma rays to Cm^{243}. -- Table 10 shows the relative abundance per alpha particle of the two highest energy

Table 10

Abundances of the Curium Gamma Rays

Sum of the abundances of the 226 and 278 kev gamma rays (determined by calculated geometry factors) relative to:	Sample whose alpha spectra are shown in Fig. 8a Experiment I	II	III	IV	Average
Cm^{242} alpha activity	1.7×10^{-4}	3.3×10^{-4}	2.6×10^{-4}	3.8×10^{-4}	3×10^{-4}
Cm^{243} alpha activity	2.9×10^{-2}	5.4×10^{-2}	4.2×10^{-2}	6.2×10^{-2}	5×10^{-2}
Cm^{244} alpha activity	3.8×10^{-2}	7.0×10^{-2}	5.5×10^{-2}	8.1×10^{-2}	6×10^{-2}

	Sample whose alpha spectrum is shown in Fig. 8d Experiment I	II	Average
Cm^{242} alpha activity	3.1×10^{-5}	2.5×10^{-5}	3×10^{-5}
Cm^{243} alpha activity	4.2×10^{-2}	3.2×10^{-2}	4×10^{-2}
Cm^{244} alpha activity	2.6×10^{-2}	2.0×10^{-2}	2×10^{-2}

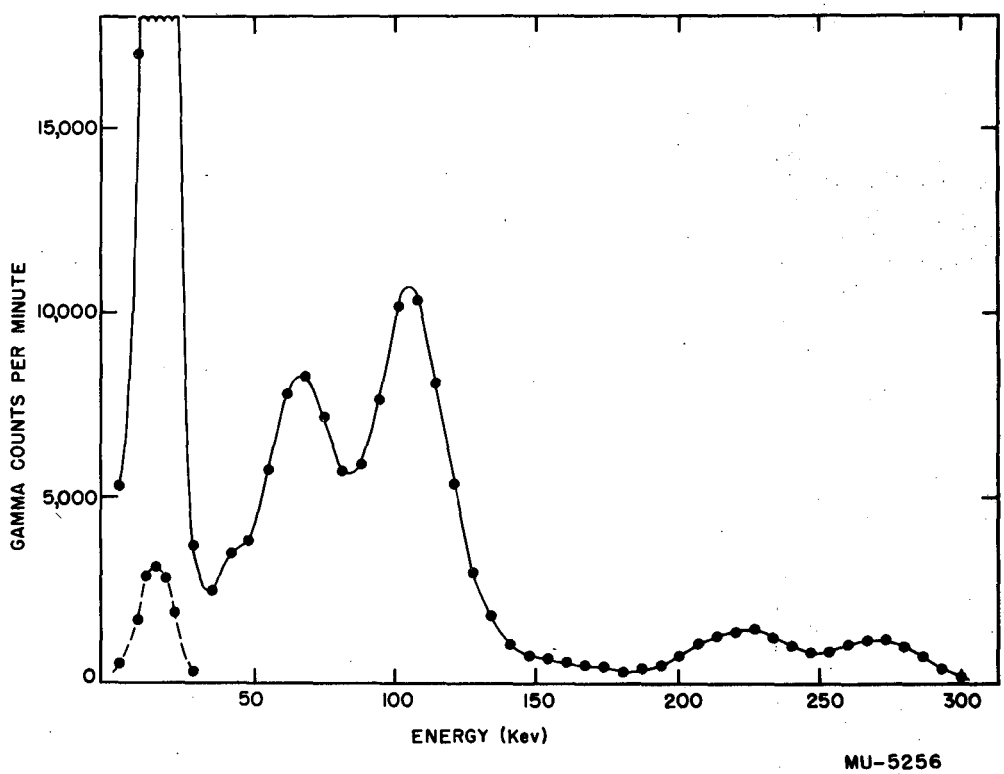


Fig. 9. Gamma spectrum of sample whose alpha spectrum is shown in Fig. 8a.

—o— Gamma counts per minute are indicated by the ordinate scale.

- -o- - Gamma counts per minute are 10 times the value indicated by the ordinate scale.

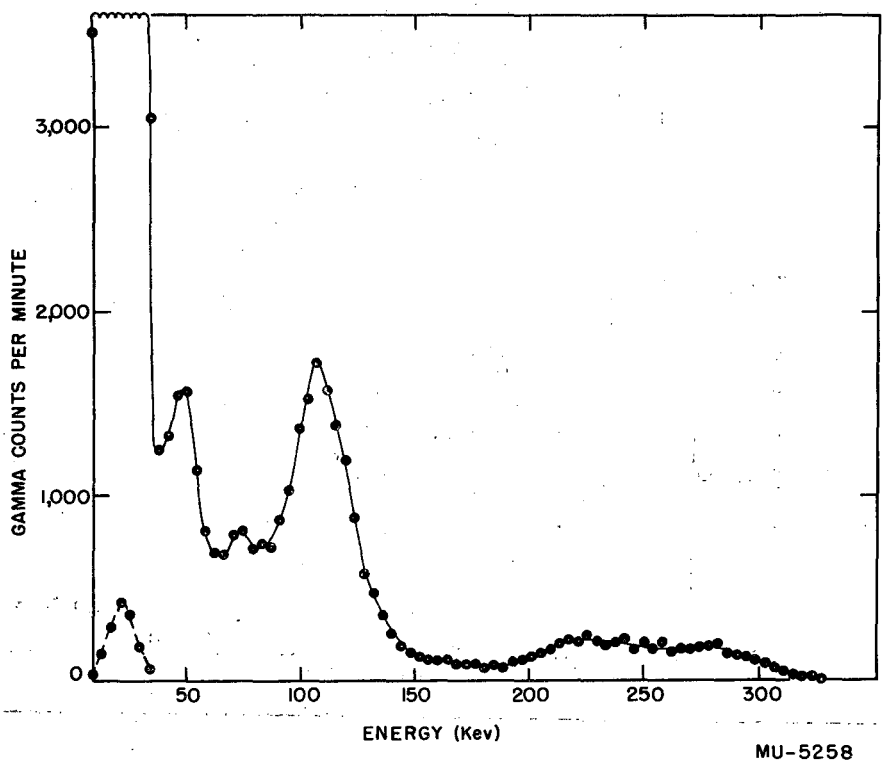


Fig. 10. Gamma spectrum of sample whose alpha spectrum is shown in Fig. 8d.

—o— Gamma counts per minute are indicated by the ordinate scale.

- -o- - Gamma counts per minute are 50 times the value indicated by the ordinate scale.

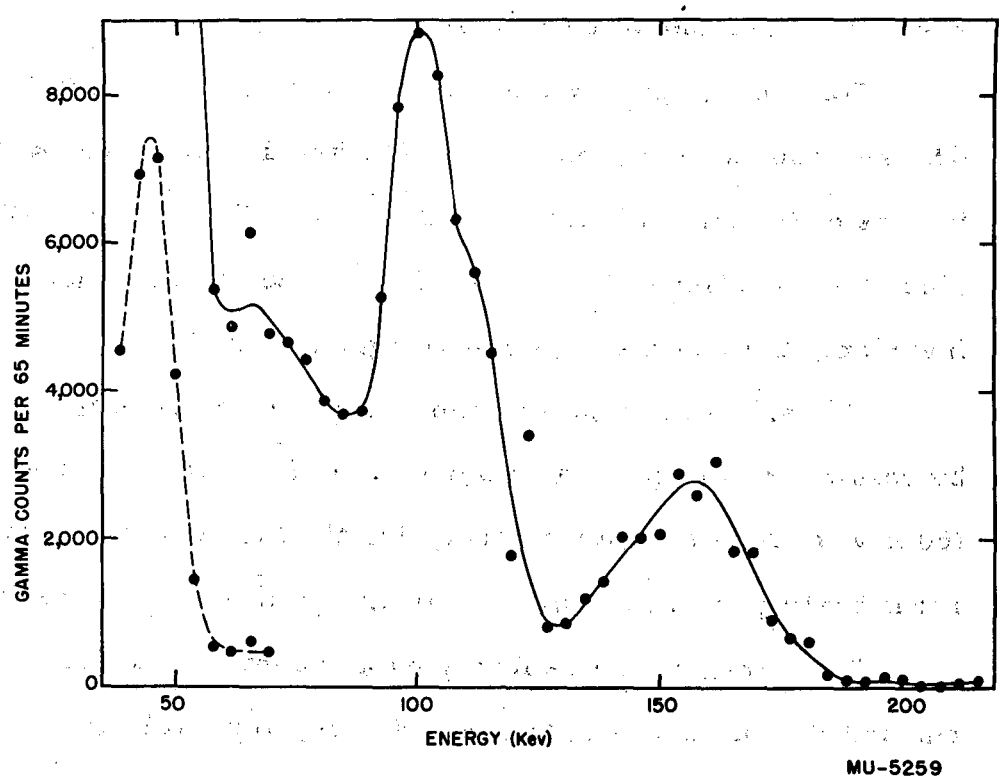


Fig. 11. Gamma spectrum of nearly isotopically pure Cm^{242}.

—o— Gamma counts are indicated by the ordinate scale.

--o-- Gamma counts are 10 times the value indicated by the ordinate scale.

gamma rays which were observed. The abundances of these gamma rays were determined by multiplying the gamma counting rate by a calculated geometry factor. This factor was dependent on the distance between the sample and crystal, the diameter of the crystal, the shape of the sample, and the fraction of the gammas striking the crystal which are registered on the scaler. Although the absolute magnitude of the numbers may be in error, their relative values should be reliable.

For the samples whose alpha spectra are shown in Fig. 8a and 8d, the abundances of the 226 and 278 kev gamma rays vary by factors of 10 and 2.7 relative to Cm^{242} and Cm^{244}, respectively. Since the abundances relative to Cm^{243}, however, remain nearly invariant, the gamma rays are assigned to Cm^{243}.

The energies of these gamma rays were determined accurately by calibrating the gamma analyzer with U^{235} (184 kev) and Am^{241} (60 kev) gamma standards, analyzing the high energy gammas, recalibrating, reanalyzing the curium gammas, and recalibrating.

By averaging the calibrations adjacent to a curium sample run and the curium sample runs adjacent to a calibration, we compensated for drift in the analyzer. In Table 11 are given the energies of the high energy gammas as a function of the calibration used.

There was a distinct change in the peak position of the standards between the first and second calibration runs and little change between the second and third. The average is taken of columns I and II, II and III, and III and IV, giving respectively

Table 11

Energies of Cm^{243} Gamma Rays

	Calibration 1 and Gamma analysis 1	Calibration 2 and Gamma analysis 1	Calibration 2 and Gamma analysis 2	Calibration 3 and Gamma analysis 2
Energy of gamma rays (kev)	219	232	223.5	225.6
	271	281	276.6	279.6

226.5 and 276, 227.7 and 278.8, and 225 and 278 kev. It is seen that after compensating for drift in the analyzer, a reasonably constant energy is obtained for the gamma rays. The energies are taken as the average of the three final values, 226 and 278 kev.

The abundance of the 104 kev radiation relative to the sum of the abundances of the 226 and 278 kev gamma rays was measured as 3.7 and 4.3 for the samples whose alpha spectra are shown in Fig. 8a and 8d, respectively. Therefore, this radiation belongs in the Cm^{243} decay scheme also, since its abundance in the two samples varies by factors of 9, 1.1 and 2.3 relative to Cm^{242}, Cm^{243} and Cm^{244}, respectively.

The average of four energy measurements of this gamma ray is 104 kev with the values ranging from 99 to 106 kev.

In order to establish a decay scheme for Cm^{243} it was desirable to determine with which alpha group the 226 and 228 kev gamma rays were in coincidence.

To effect this, the photographic camera on the receiver of the spectrograph was replaced with a zinc sulfide phosphor mounted on a lucite light pipe. This phosphor was masked except for a 1/8 in. x 2 in. slit. Behind the light pipe was a 5819 RCA photomultiplier tube. The sodium iodide detector was mounted directly behind the spectrograph source, the vacuum being maintained in the spectrograph by means of a 10 mil aluminum window between the source and detector.

The energy of alpha particles arriving at the slit would be proportional to the square of the field. This is determined from equation (3) at constant radius. The sample whose alpha spectra

are shown in Fig. 8a and whose gamma spectra are shown in Fig. 9 was mounted in the spectrograph and coincidences were determined between radiation in the K x-ray region and various values of the magnetic field corresponding to various alpha particle energies. The alpha particle energies for the various magnetic field settings were determined by taking the energy of α_0 of the Cm^{242} peak as 6.110 Mev. A graph of the coincidences is shown in Fig. 12. Marked on the graph are the positions of the Cm^{243} and Cm^{244} peaks. It is seen that at least the bulk of the radiation is in coincidence with the 5.777 Mev alpha group of Cm^{243}. Setting the magnetic field so that the alpha particles striking the receiver slit corresponded to the maximum of the alpha-gamma coincidences curve shown in Fig. 12, coincidences were run between these alpha particles and the gamma spectrum around 226 and 278 kev with the single channel gamma analyzer. The results are shown in Fig. 13. The runs varied in length of time from 1 to 16 hours. From abundance measurements one would not expect these gamma rays to be in coincidence with a tail from the 5.732 Mev Cm^{243} alpha group. Therefore, these gamma rays are in coincidence with the 5.777 Mev group.

2. Abundances of the Cm^{243} gamma rays. -- As stated previously, the absolute magnitude of the abundance of the 226 and 278 kev gamma rays may be in error. In order to check the geometric calculations, a sample of Am^{241} was gamma analyzed and the abundance of the 60 kev gamma ray, calculated with the appropriate geometry factor, was found to be a factor of 4 lower than the value of 0.40 gamma ray per alpha disintegration given by Beling, Newton and Rose.[6] Therefore, all the abundances measured

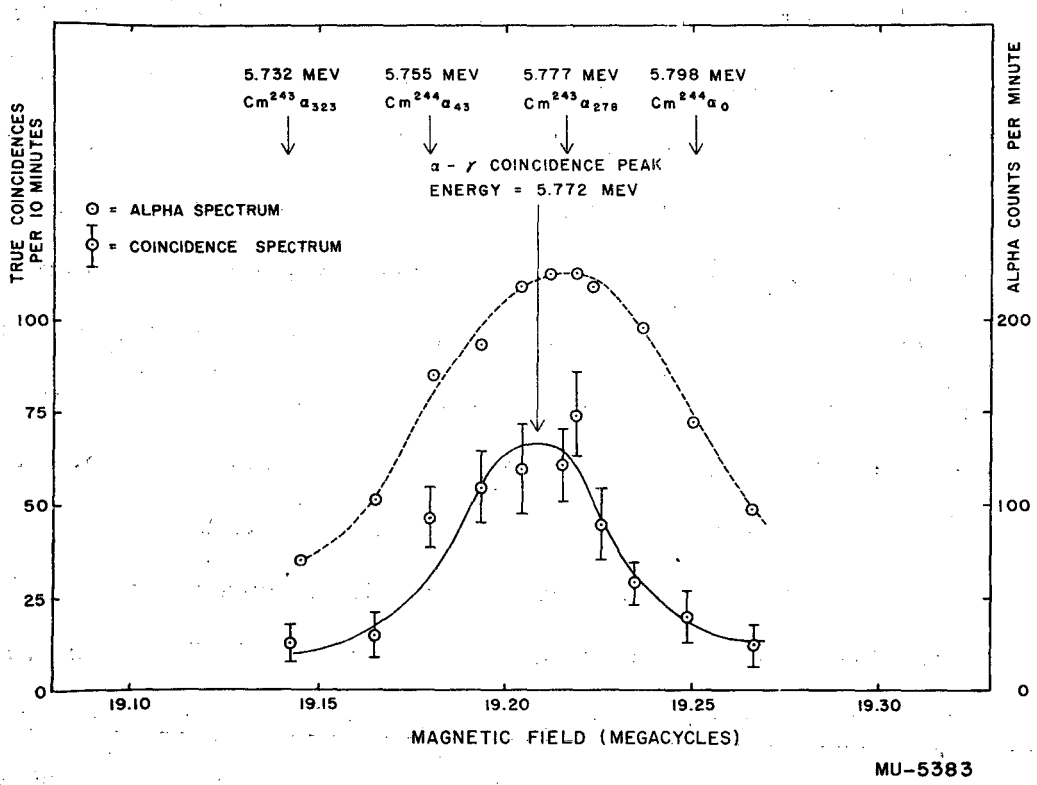


Fig. 12. Alpha-K x-ray coincidences of Cm^{243}.

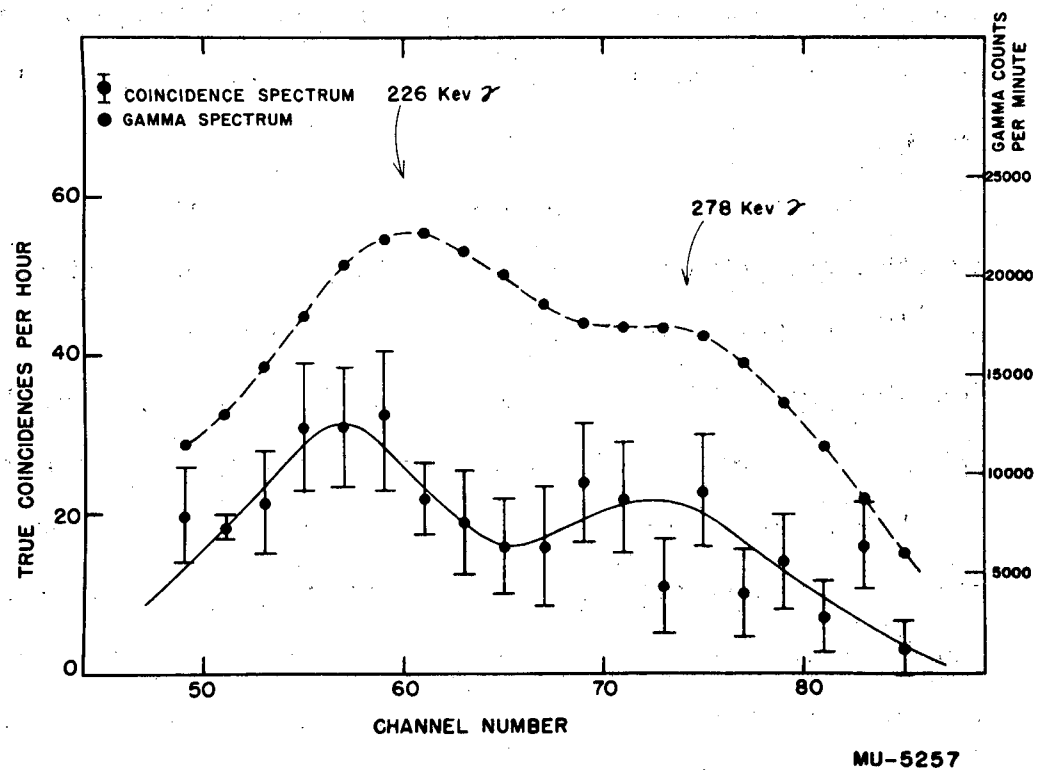


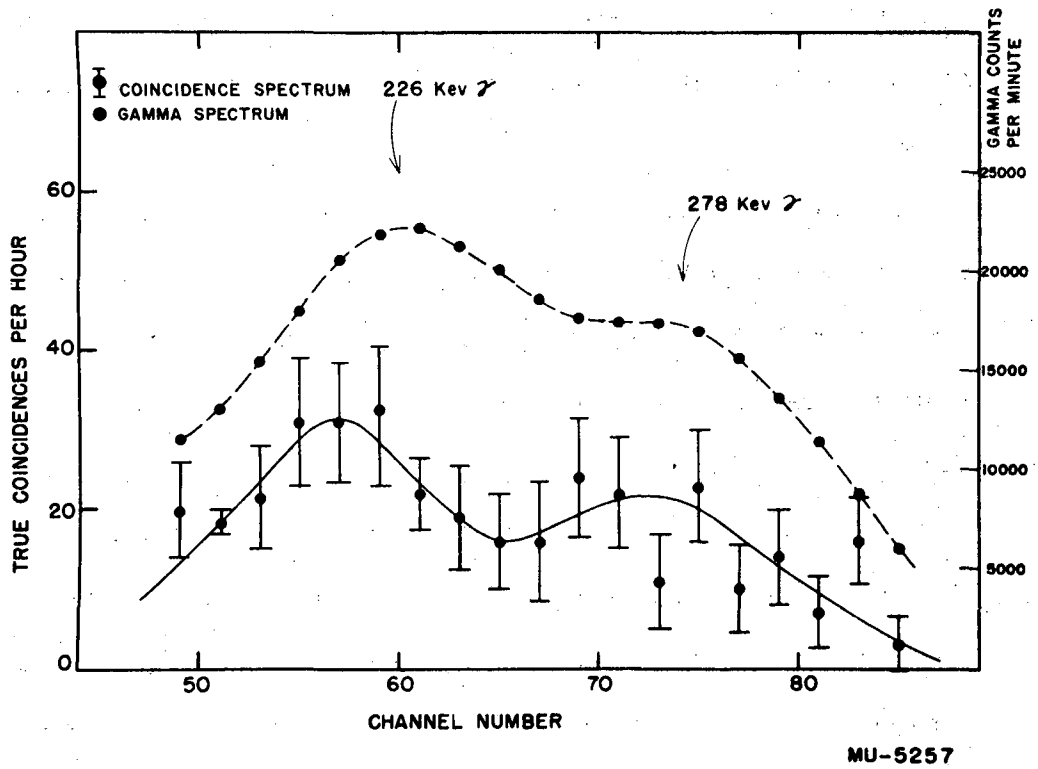
Fig. 13. Alpha-gamma coincidences of Cm^{243}.

before must be multiplied by a factor of 4. Thus from Table 10, the abundance of the 226 plus and 278 kev gamma is $4 \times 4.2 \times 10^{-2}$, or approximately 17 percent of the Cm^{243} alpha disintegrations.

In order to obtain more accurate absolute abundance measurements, the NaI crystal was repacked with MgO in its copper case. Upon analyzing a sample of Am^{241} again, the abundance of the 60 kev gamma ray, calculated with the appropriate geometry factor, was found to be about 40 percent too high. The reason for the large apparent increase in geometry upon repacking the crystal was due to the removal of MgO from between the crystal and photomultiplier faces.

More consistent results were obtained by determining the effective geometry experimentally. This was done by placing an Am^{241} sample of known alpha activity in exactly the same position occupied previously by the curium sample. Then by using the value 0.40 gamma ray per alpha disintegration, we determined the effective geometry of the arrangement. Abundances of the 104 kev radiation obtained before and after repacking the crystal were 64 and 56 percent, respectively, relative to the Cm^{243} alpha activity. The average of these two values, 60 percent, is taken as the best value.

The sum of the abundances of the 226 and 278 kev gamma rays is about 1/4 the abundance of the 104 kev gamma rays, as was mentioned in a previous section. Therefore, their intensity would comprise about 15 percent of the Cm^{243} alpha transitions in fair agreement with the value of 17 percent obtained in the previous section.



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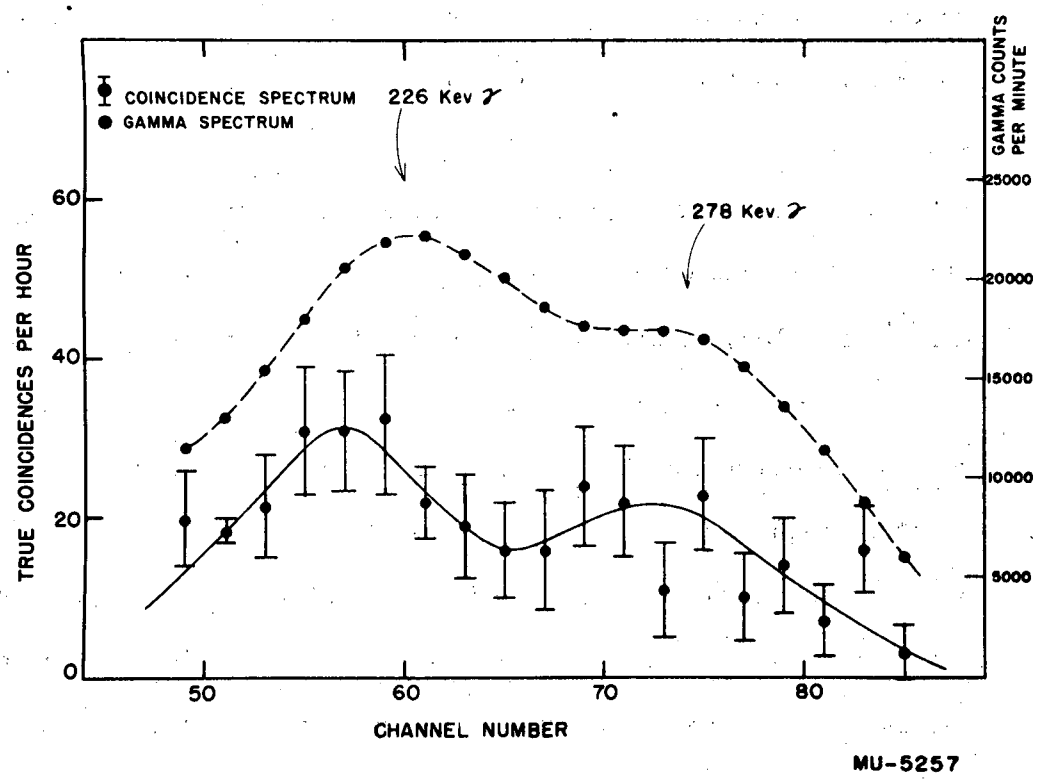

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There have been no corrections, however, for the change in counting efficiency of the NaI crystal with energy. A rough calculation with the experimentally determined counting efficiency of the Cs^{137} 662 kev[13] gamma ray indicated that the average counting efficiency of the 226 and 278 kev gamma rays would be about 50 percent, while the counting efficiency of the gamma rays below 150 kev would be above 98 percent. Therefore, the best value for the sum of the abundances of the 226 and 278 kev gamma rays is 2 x 15 percent = 30 percent of the Cm^{243} alpha disintegrations.

The relative abundance of the 226 to the 278 kev gamma ray cannot be determined very accurately because of the difficulties in resolution. The best value for the ratio appears to be 55:45.

3. Assignment of gamma rays to Cm^{242}. --Since a 44 kev gamma ray appears in the sample whose alpha spectra are shown in Fig. 8d and is not seen in Fig. 8a, it is readily assignable to Cm^{242}. This assignment is verified by the appearance of the 44 kev gamma ray in the sample of Cm^{242} with high isotopic purity (Fig. 11). The energy of this gamma ray is the average of seven measurements with the values ranging from 40 to 49 kev.

Other workers in this laboratory have found evidence of a 44 kev transition in the decay of Cm^{242}. Ghiorso[9] has observed the gamma ray with a proportional counter coupled to a pulse height analyzer and Martin[14] has measured it with a scintillation counter spectrometer. O'Kelley[15] has measured a series of conversion lines corresponding to a gamma ray of about 43 kev and Dunlavey and Seaborg[16] have observed L and M electrons corresponding to a 45 kev gamma ray in coincidence with 23 percent of the alpha particles.

The 100 kev gamma observed in Fig. 11 is too intense to be due to Cm^{243}. From Fig. 9 the maximum intensity of the Cm^{243} 104 kev radiation expected from the maximum intensity of the Cm^{243} high energy gamma rays would be about 1/6 of the observed abundance. Therefore, this gamma ray is assigned to Cm^{242}. The energy of this was taken as the average of four values ranging from 99 to 102 kev.

The 157 kev gamma observed in Fig. 11 is not seen in Fig. 9 or Fig. 10 and is therefore assigned to Cm^{242}. The energy of this gamma ray was taken as the average of four values which varied from 155 to 159 kev.

4. Abundances of the Cm^{242} gamma rays. -- The abundance of the 44 kev gamma ray is 0.041 percent of the Cm^{242} alpha activity. The geometry of the system was determined with the Am^{241} 60 kev gamma ray assuming 0.40 gamma ray[6] per alpha particle.

The abundance of the 100 kev gamma ray was measured as 18 percent of the 44 kev gamma ray, or 0.0073 percent of the Cm^{242} alpha activity. This value must be corrected for the slight amount of Cm^{243} in the sample. From Fig. 9 and 10 it is seen that the ratio of the peak heights of the Cm^{243} 104 kev radiation to the 226 kev gamma ray is about 10:1. Applying this ratio to low intensity Cm^{243} gamma rays which were detected in the Cm^{242} sample, we find the Cm^{243} 104 kev contribution is 19 percent of the observed 100 kev radiation. Therefore, the corrected abundance of the 100 kev gamma ray is 0.006 percent of the Cm^{242} alpha activity.

The abundance of the 157 kev gamma ray was measured as

6.6 percent of the 44 kev gamma ray, or 0.0027 percent of the Cm^{242} alpha activity. This value has not been corrected for the change of counting efficiency of the NaI crystal with energy. The counting efficiency at 157 kev is expected to be substantially the same as at lower energies.

5. Miscellaneous gamma rays. -- The 60 kev gamma ray observed in Fig. 9 is probably due to Am^{241} since a mass spectrographic analysis showed a considerable amount of Am^{241}. The 70 kev gamma rays observed in Fig. 10 and 11 are probably the escape peaks of the 104 and 100 kev radiations. The escape peak is caused by the escape of iodine K electrons from the NaI crystal, thus causing a sattelite gamma ray lower in energy than the principal gamma ray. This energy separation is equivalent to the K binding energy of the iodine K electron (33 kev).

6. Decay scheme of Cm^{244}. -- The decay scheme consists only of the alpha spectrograph data as shown in Fig. 14a.

7. Decay scheme of Cm^{243}. --The 104 kev radiation in Cm^{243} alpha decay could be due to a gamma ray, K x-rays, or both. If one takes the energies and abundances of the K x-rays observed by Jaffe, Browne and Perlman,[17] and calculates a weighted average, the resulting plutonium K x-ray energy is 103.8 kev in excellent, but probably fortuitous, agreement with our value. The decay scheme, deduced from only the alpha and gamma data on Cm^{243}, is shown in Fig. 14b. It is interesting to examine the data on the beta spectra of Np^{239} investigated by other workers. Beta particles of 0.718, 0.655, 0.441, 0.380 and 0.329 Mev[18] have been observed. Conversion

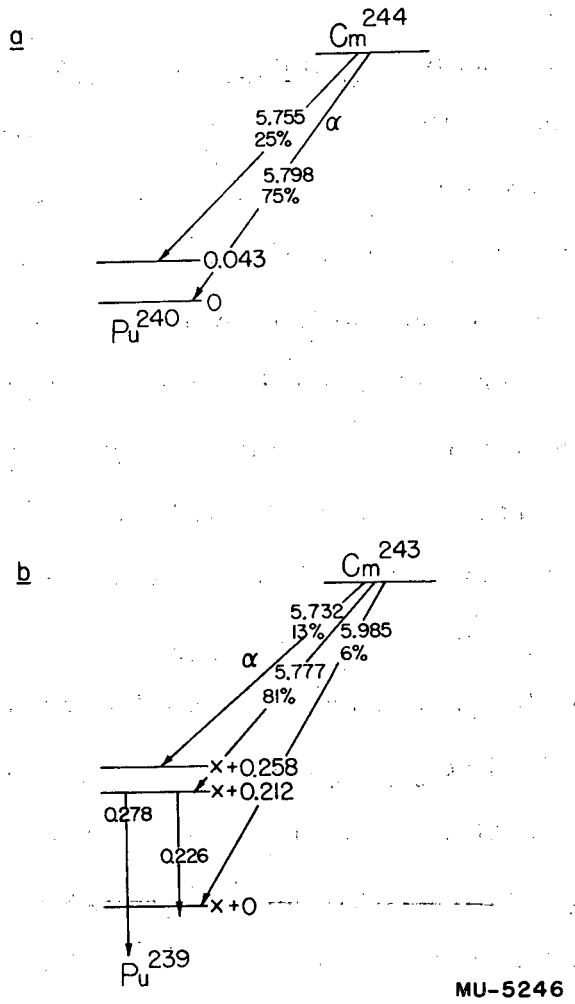

Fig. 14. a) Cm^{244} decay scheme.

b) Cm^{243} decay scheme.

lines corresponding to gamma rays with energies of 0.013, 0.019, 0.044, 0.049, 0.057, 0.061, 0.067, 0.077, 0.105, 0.209, 0.228, 0.254, 0.277, 0.285, 0.316, 0.334 Mev[18] have also been observed. Conversion lines of gamma rays with energies of 0.210, 0.227, and 0.276 Mev were found to be in coincidence with a 0.435 Mev beta particle.[19] The 0.210 Mev gamma ray is in coincidence with the 0.067 Mev gamma ray, but not with the 0.276 kev gamma ray.[19]

Using our knowledge of the levels in Pu^{239} deduced from the Cm^{243} alpha decay, we obtain the energy level scheme for Pu^{239} shown in Fig. 15.

It is interesting to investigate the multipole order of the 226 and 278 kev gamma rays observed in Cm^{243} alpha decay. These gamma rays are emitted by the Pu^{239} excited state 277 kev above the ground state. The values of the energy levels given in Fig. 15 are those determined by beta decay. According to Bell and Graham,[19] this state is metastable with a half-life of 1.1×10^{-9} seconds. From the decay scheme shown in Fig. 15 it is evident that over 80% of the Cm^{243} disintegrations must pass through the 277 kev level. Since the 226 and 278 kev gamma rays comprise 30 percent of the Cm^{243} disintegrations, the half-life for gamma emission will be

$$\frac{1.1 \times 10^{-9} \text{ sec}}{\approx 0.3} \approx 4 \times 10^{-9} \text{ sec.}$$

For each gamma ray then, the half-life will be about 10^{-8} seconds.

Using the form of Weisskopf's lifetime relations given by Goldhaber and Sunyar,[20] we would expect lifetimes of the order of 10^4 and 10^6 seconds, respectively, for electric and magnetic 2^4 pole

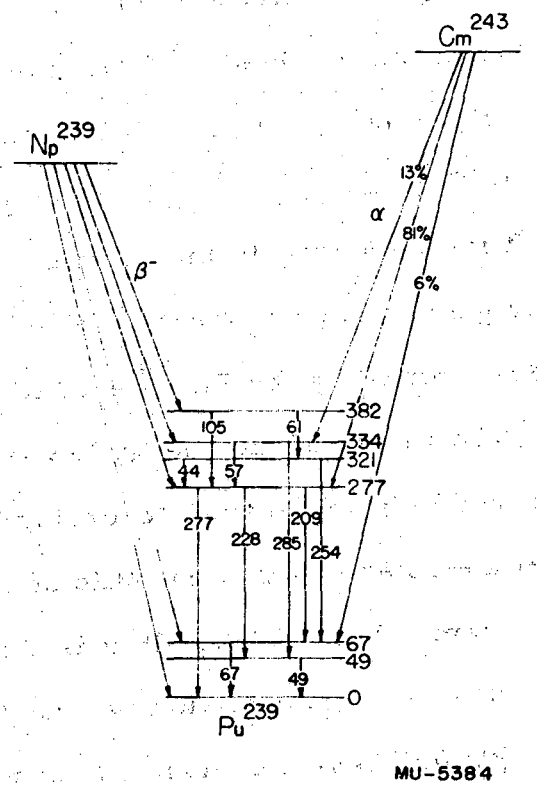

Fig. 15. Cm^{243}-Np^{239} decay scheme.

radiation. Since these values differ by at least a factor of 10^{12} from the experimental value, it is safe to conclude that the 226 and 278 kev gamma transitions are either electric or magnetic dipole, electric or magnetic quadrupole, or electric or magnetic octopole.

This difficulty could be resolved if the conversion coefficients of the gamma rays were known. These conversion coefficients can be calculated if the relative abundances of the conversion lines are known. These relative abundances were obtained* and the conversion coefficients are shown in Table 12. From the data in this table, both the 226 and the 278 kev transitions appear to be magnetic dipole radiation.

8. Decay scheme of Cm^{242}. -- The 44 kev gamma ray very probably corresponds to the transition between the levels in Pu^{238} populated by α_{44} and α_0 of Cm^{242}. The conversion coefficient of this gamma ray, assuming no other transitions take place between the two levels, would be 26.3%/0.041% ≅ 640. The multipole order of the transition can be determined providing the L conversion coefficients can be found. Dunlavey and Seaborg,[16] in their study of alpha electron coincidences in photographic plates, found that 83 percent of the coincidences were from L conversion electrons. Therefore, the L conversion coefficient for the 44 kev gamma ray would be $640 \times 0.83 = 5.3 \times 10^2$. From Gellman's tables,[22] we would expect theoretical L conversion coefficients of 1.5, 6×10 and 6×10^2 for

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*I would like to express my appreciation to M. S. Freedman for sending to me the results communicated to him by Harry Fulbright.

Table 12

Multipole Order of Cm^{243} Gamma Rays

Energy of gamma ray (kev)	Experimental α_K	Theoretical K conversion coefficients from Rose's tables.[21]				Experimental α_L	Theoretical L conversion coefficients from Gellman's tables.[22]		
		E_1	E_2	M_1	M_2		E_1	E_2	M_1
226	1.8 → 3.6	0.06	0.14	4.1	8.7	0.4 → 0.8	0.015	0.35	0.72
278	1.8 → 3.5	0.04	0.1	2.3	4.6	0.3 → 0.7	0.007	0.15	0.36

Energy of gamma ray (kev)	Experimental K/L ratio	Empirical K/L ratios from Goldhaber and Sunyar.[20]				Experimental $L_1:L_2:L_3$ conversion ratio.[23]	Theoretical $L_1:L_2:L_3$ ratios from Gellman's tables[22]		
		E_2	E_3	M_1	M_2		E_1	E_2	M_1
226	4.3	0.6	0.3	7.6	5.2	1:0.09:0.009	1:0.39:0.28	1:3.9:1.6	1:0.1:0.001
278	5.3	0.8	0.4	7.7	5.7	1:?:0.006	1:0.32:0.2	1:3.3:1.5	1:0.11:0.001

Energy of gamma ray (kev)	Experimental half-life (sec)	Mean life calculated from Weisskopf's formula (seconds)[24]					
		E_1	E_2	E_3	M_1	M_2	M_3
226	10^{-8}	10^{-14}	6×10^{-4}	5×10^{-3}	10^{-12}	10^{-6}	1
278	10^{-8}	10^{-14}	2×10^{-9}	10^{-3}	10^{-12}	5×10^{-7}	1

E_1, M_1 and E_2 radiation, respectively. Therefore, the 44 kev radiation is electric quadrupole and hence the first excited state of Pu^{238} has a spin of 2 even parity. This result is in agreement with the general principle stated by Goldhaber and Sunyar that the spin of the first excited state of an even-even nucleus is 2 even parity.

The 100 kev radiation could be a gamma ray corresponding to the transition between the levels in Pu^{238} populated by α_{148} and α_{44} or it could be a K x-ray.

One can choose between the gamma ray and the K x-ray assignment by considering selection rules which apply to alpha decay theory. These rules are developed in Appendix 1. These alpha decay selection rules indicate that all levels in an even-even nucleus which are populated by alpha decay have odd spin and odd parity or even spin and even parity. In addition, all transitions to the ground state will have an electric multipole order.

If the 100 kev radiation was a K x-ray of a transition between the levels in Pu^{238} populated by α_{148} and α_o of Cm^{242}, the transition would have to be electric. Since the maximum abundance of any gamma ray in Cm^{242} above 100 kev and below 300 kev is 1/2.7 times the abundance of the 100 kev radiation, the minimum K conversion coefficient would be 2.7. From Rose's tables[21] of theoretical K conversion coefficients, the multipole order of the transition would be E_5. From Weisskopf's[24] formula, the expected half-life would be some 10,000 years. Since the 100 kev radiation is seen soon after the curium is purified, this radiation is not a K x-ray of any transition to the ground state of Pu^{238}.

If the 100 kev radiation were a K x-ray of the 157 kev transition between two excited levels of Pu^{238}, the multipole order would have to be magnetic in order to have a reasonable lifetime for the gamma emission. From Rose's tables,[21] the theoretical K conversion coefficient for magnetic transitions is at least 8 in contrast to the experimental value of 2.7. Therefore, the bulk of the 100 kev radiation is not a K x-ray of the 157 kev transition but is probably a gamma ray. If this gamma ray corresponds to the transition from the levels in Pu^{238} populated by α_{148} and α_{44} of Cm^{242} and the 148 level is not de-excited by any other transition, the conversion coefficient of this gamma ray is (0.035 - 0.006)/0.006 = 5. The ratio of M to L conversion is usually small for gamma energies of 50 kev or higher. So the total conversion coefficient is equal to the L conversion coefficient within the experimental error involved in the calculations. From Gellman's tables,[22] one would expect theoretical conversion coefficients of 0.1, 4, and 7, respectively, for E_1, M_1 and E_2 multipole orders. Therefore, this 100 kev gamma ray would correspond to an M_1 or an E_2 transition. The spin of the 148 kev level in Pu^{238} would then be 4 even parity, 2 even parity, or 0 even parity.

The decay scheme for Cm^{242} is given in Fig. 16a. The 157 kev gamma ray is not included as there is no experimental evidence showing where it belongs. A possible place in the decay scheme will be mentioned in the section on empirical correlations.

Some of the Pu^{238} levels populated by alpha decay of Cm^{242} are also populated by Np^{238} decay. Beta particles of 1.272 Mev

(47 percent) and 0.258 Mev (53 percent) as well as conversion lines of 20.8, 24.7, 37.4, 41.9, 80.2, 97.9, 859 and 913 kev have been seen.[25] Coincidences were observed between L x-rays and soft and hard beta particles, L x-rays and L x-rays, soft beta particles and hard gamma rays, L x-rays and hard gamma rays, and hard beta particles and 100 kev gamma rays.[25]

The conversion lines of 20.8, 24.7, 37.4 and 41.9 correspond to the expected energies for L_{II}, L_{III}, M_I, and N_I conversion of a 43.3 kev gamma ray and the 80.2 and 97.9 kev electrons correspond to the expected energies for L and M conversion of a 103 kev gamma.[26]

A decay scheme which we postulated in private communication[27] is shown in Fig. 16b.

The intensity of the electrons of the highly converted 100 kev gamma ray, however, is a factor of 20 less than the intensity of the electrons corresponding to the 44 kev gamma ray. So this decay scheme is unlikely.

A decay scheme which satisfies all the known experimental data is shown in Fig. 17. This decay scheme assumes a 1.168 Mev beta particle in 3 percent abundance, in order to account for the coincidences observed between the hard beta particles and the 100 kev gamma rays. The 0.258 Mev beta particle has an allowed shape and a log (ft) value of 6.2. According to Nordheim's[28] selection rules, the spin and parity change should be 1 No or 0 Yes. The 1.272 Mev beta particle has an allowed shape and a log (ft) value of 8.5. Therefore, according to Nordheim's selection rules, the spin change should also be 1 No or 0 Yes. From simple considerations

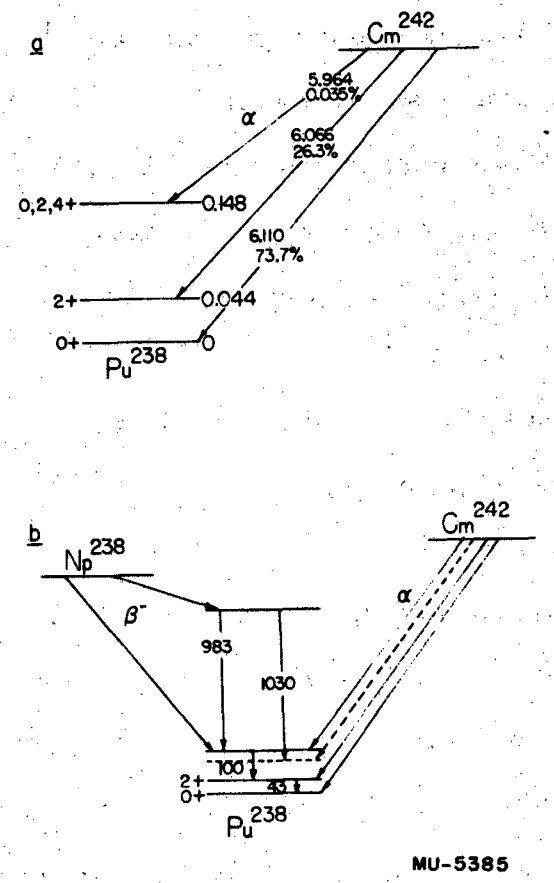


Fig. 16. a) Cm^{242} decay scheme.

b) Untenable Cm^{242}-Np^{238} decay scheme.

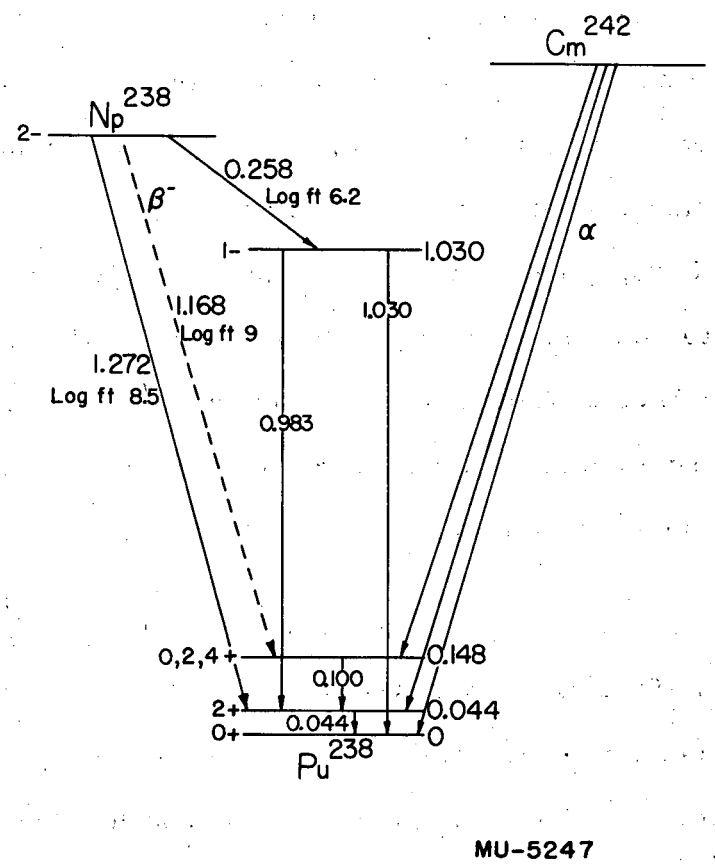


Fig. 17. Cm^{242}-Np^{238} decay scheme.

of shell theory, the parity of the ground state of Np^{238} should be negative. Since there must be a parity change then in the 1.272 Mev beta emission, the spin change is 0 Yes. So the spin of Np^{238} is 2 minus.

Since the 1030 kev level decays to the states with spins of 0 and 2, its spin must be 1 or 2. Therefore, taking into account the selection rules affecting the 0.258 Mev beta particle, the spin of the 1030 kev level is 2 even parity or 1 odd parity. The K-conversion coefficients of both hard gamma rays are about 1 percent, which from Rose's tables[21] correspond to E_1 transitions. Therefore, the spin of the 1030 kev level is 1 odd parity.

If the 148 kev level had a spin of 0 even parity, one might expect to see a gamma ray corresponding to the transition from the 1030 kev level. Goldhaber and Sunyar,[20] however, state that no quantitative prediction can be made as to the relative intensities of competing electric transitions.

E. Complex Alpha Spectrum of Am^{241}

Before discussing the complex structure of Am^{241}, the energy determination will be mentioned so that the groups may be referred to according to energy. The nuclide Am^{241} was that by which the element was first identified,[29] and it is prepared in isotopically pure form from Pu^{241} decay.[30] The half-life is given as 470 years[31] and 475 years[32] which corresponds to a specific alpha activity of 6.95×10^6 disintegrations per minute per microgram. Previous to the present studies the measurement of alpha-energy has been made with an ionization chamber coupled to

a pulse-height discriminator from which the value 5.48 Mev was reported.[33] Although one could observe from the pulse-height analysis that the spectrum was not simple, it was not possible to resolve it into its components. The measurement therefore gives the energy of the principal group distorted to an unknown extent by one or more other groups.

The energy for Am^{241} was determined with the spectrograph by comparing that of the principal group with two standards, Po^{210} and Em^{222}. The other groups of Am^{241} were assigned energies by comparison with the main group.

The comparison between Am^{241} and Em^{222} is shown in Fig. 18. The plate was made by placing sources of Am^{241} and radium simultaneously in the spectrograph for an exposure of 27 hours. That the higher energy group is that of Em^{222} could be proved by comparing the observed number of tracks with that expected from the observed Po^{218} tracks caught on another part of the plate. If it was assumed that half of Po^{218} formed from Em^{222} left the source by the recoil mechanism, the observed number of tracks in the high energy component was in agreement with the expected amount from Em^{222}. The resolution of the curve in Fig. 18 was aided by a separate exposure with the radium source alone in which the width of the Em^{222} peak at half-maximum was determined. The distance between the peaks was 2.7 ± 0.3 mm which corresponds to an energy difference of 11 ± 2 kev. Taking the energy of Em^{222} as 5.486 Mev,[2] the energy of the principal group of Am^{241} becomes 5.475 ± 0.002 Mev.

Similar exposures were made with Po^{210}, the results of which are shown in Fig. 19. The distance between the peaks was

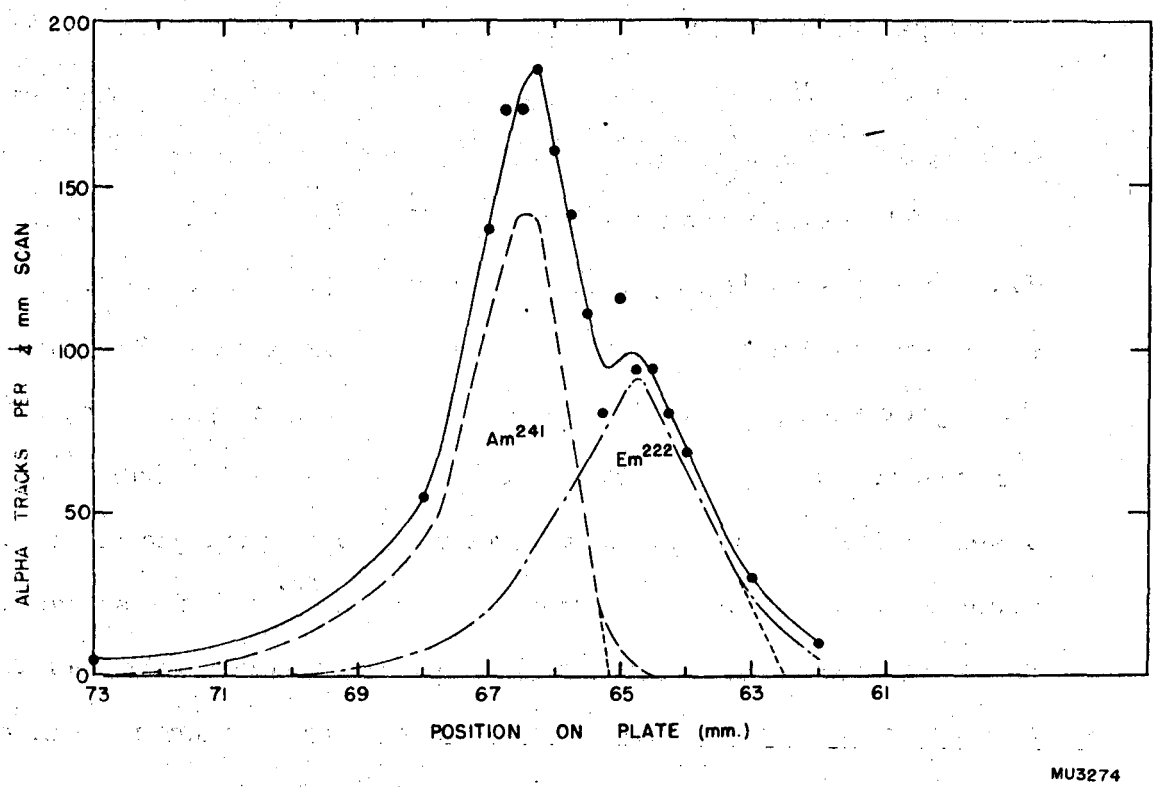


Fig. 18. Am^{241} - Em^{222} energy comparison.

Dispersion = 3.96 kev/mm.

——— spectrum as measured.

—— - —— Em^{222} peak resolved by subtracting background and using half-width for the peak as determined on a separate measurement with the Em^{222} sample alone.

—— —— resolved alpha group of Am^{241}.

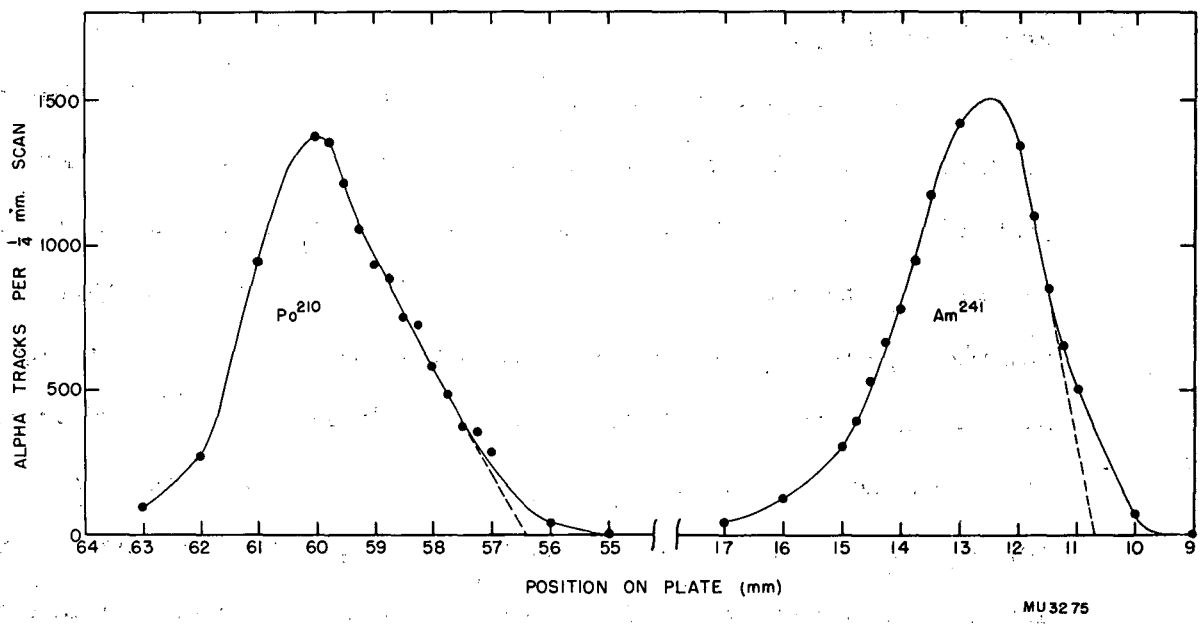


Fig. 19. Am^{241}-Po^{210} energy comparison.

Dispersion = 3.90 kev/mm.

45.8 mm which corresponds to an energy difference of 179 ± 2 kev. From this measurement the main group of Am^{241} has an energy of 5.477 ± 0.002 Mev. The energy which we shall use is 5.476 ± 0.002 Mev. As will be described, the most energetic alpha group is higher in energy than this group by 70 kev, therefore its energy is 5.546 Mev. The decay energy of Am^{241} is accordingly 5.640 Mev.

There are now known to be six measurable alpha groups of Am^{241},[34] three of which are of greater energy than the most prominent group.

The complete alpha-spectrum is shown in Fig. 20. For these particular data, the source consisted of 2.9 micrograms of Am^{241} (2.0×10^7 disintegrations/minute) and the exposure was for 94 hours. Because of the disparity in abundance of the different groups, complete peaks cannot be shown to the same scale. In exposing the plate long enough to register a statistically significant number of tracks of the rare groups, too many tracks for counting were registered at the positions of the principal groups. Partial scans across the plate were made for these peaks and the results are also shown in Fig. 20. The peak widths at half-maximum are essentially the same for all peaks.

The relative abundances of the groups were obtained by counting the tracks of the two most prominent groups on a plate exposed for a shorter period of time and comparing with the rarer peaks from the long exposure. The abundances were virtually the same whether integrated numbers of tracks under the peaks or the

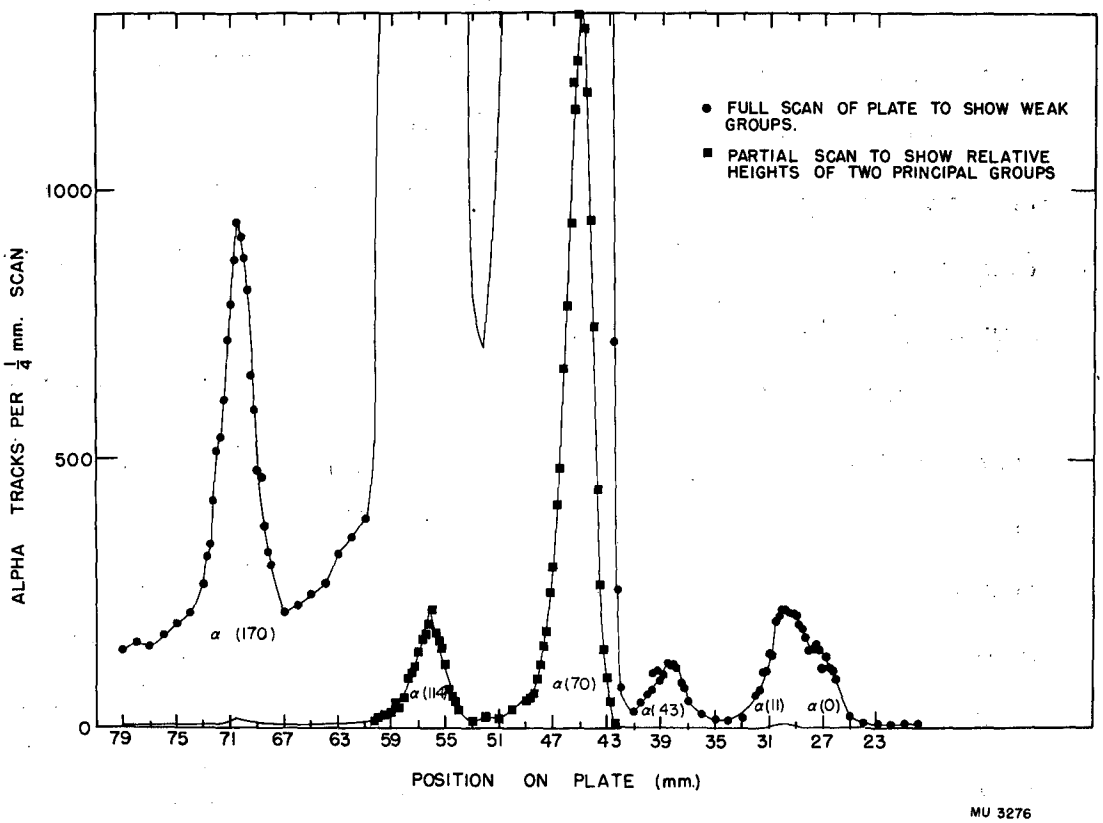


Fig. 20. Am^{241} complete alpha spectrum.

peak heights were compared. A summary of the abundances and the corresponding partial half-lives which will be referred to later are given in Table 13. The values in each case represent at least two independent measurements. The sum in abundance of the two highest energy groups is known with better precision than each group separately because of the uncertainty in resolution. For this reason they are listed both ways in Table 13.

The energy range of the observed peaks was 5.38 to 5.55 Mev. Counting the plate outside of this range revealed that there can be no peak between 5.21 and 5.38 Mev of greater abundance than 0.17 percent, and from 5.55 to 5.64 Mev none in more than 0.07 percent abundance. These limits were based on the respective background counts in the regions.

Determinations of energies of the several groups were made relative to the principal alpha group, α_{71}, which was standardized against Em^{222} and Po^{210} as already described. The actual comparisons were made by extrapolating the high energy side of each peak to the intercept after subtraction of the estimated background count and tailing from other groups. The method for obtaining the dispersion in order to translate positions on the plate to energy differences has already been described.

The results of several measurements of alpha particle energies are summarized in Table 14. The measured alpha energy differences with their estimated limits of error are as indicated. Also shown are the selected best values and the corresponding energies of the alpha groups based on 5.476 Mev for the most prominent group. In the last column are shown the energy levels above the ground state

Table 13

Abundances of Am^{241} Alpha Groups

Alpha Group[a]	Percentage Abundance	Partial Half-life[b] (yr)
$\alpha_{(0)} + \alpha_{(11)}$	0.57 (±0.06)	
$\alpha_{(0)}$	0.23 (±0.06)	2.1×10^5
$\alpha_{(11)}$	0.34 (±0.06)	1.4×10^5
$\alpha_{(43)}$	0.21 (±0.02)	2.3×10^5
$\alpha_{(71)}$	84.2 (±1.5)	564
$\alpha_{(114)}$	13.6 (±1.4)	3500
$\alpha_{(170)}$	1.42 (±0.15)	3.3×10^4

[a]The highest energy group, designated $\alpha_{(0)}$, is taken to represent the ground-state transition. The parenthesis-enclosed figures used for the other groups indicate the energy levels in kilovolts above the ground state with which the alpha-groups are associated.

[b]Based on 475-year measured half-life.

Table 14. Energies of Am^{241} Alpha Groups.

Alpha group	Experiment number						Best values	Alpha particle energies (Mev)	Energy levels above the ground state
	33	48	59a[a]	59b[a]	61a	61b			
α(0)								5.546	0
		10.3 ±2.0	9.3 ±1.1	10.8 ±0.8	12.4 ±1.6	10.5 ±1.5	10.6 ±1.4		
α(11)								5.535	10.8
			32.4 ±0.8			31.7 ±0.7	32.0 ±0.7		
					30.8 ±1.5				
α(43)				58.6 ±1.4	59.5 ±1.3			5.503	43.4
			26.5 ±0.8				59.0 ±1.3		
						26.2 ±0.5			
α(71)								5.476 ±0.002	70.8
	42.6 ±1.1		42.4 ±0.8	42.1 ±0.8	43.2 ±1.7		42.7 ±1.1		
α(114)		95.6 ±3.6				96.5 ±2.5	97.1 ±2.5	5.433	114.1
			99 ±2						
α(170)								5.379	169.6

[a]Series a and b refer to independent counts of the alpha tracks on the same plate.

of Np^{237} with which each alpha group is associated. These levels are obtained by correcting the differences of alpha group energies for the corresponding differences in alpha decay recoil energy. It is differences between these numbers which should correspond to gamma ray energies.

1. Decay scheme of Am^{241}. --The complexity of the decay scheme of Am^{241} may be visualized readily from the number of observed levels. The presence of a 60 kev gamma ray in Am^{241} decay has been abundantly confirmed by measurements with proportional counter spectrometers,[6, 35] beta ray spectrometers,[16, 35] a bent crystal spectrometer,[36] and NaI (Tl) activated crystal spectrometers.[14, 35] Those values with reported limits of error were 59.78 ± 0.04 kev[36, 37] and 59.7 ± 0.3 kev.[6] The abundance of the gamma quantum has been reported as 40 ± 1.5 percent,[6] and 32 percent.[38] Dunlavey and Seaborg[16] report 44 percent of the Am^{241} alpha particles have no visible electrons in coincidence.

That the 60 kev gamma ray belongs in the Np^{237} is verified by alpha-gamma coincidence measurements[38, 39] on Am^{241} and by its presence in the beta decay of U^{237} [40, 41, 36] and the electron capture of Pu^{237}.[42]

The multipole order of the 60 kev gamma ray has been reported as E_1[39] from the experimental conversion coefficient and as an M_1-E_2[36] mixture from the ratios of L vacancies and the L_1 conversion coefficient.

Another abundantly confirmed gamma ray has been reported

as 26.43 ± 0.03 kev,[36] 26.3 ± 0.2 kev,[6] and 26.4 kev.[35] The respective abundances relative to the 60 kev gamma ray were 31 percent, 7.5 ± 0.8 percent, and 25 percent. The multipole order of this transition has been reported as E_1.[39]

A gamma ray of 33.36 ± 0.03 kev[7, 36] was observed with a bent crystal spectrometer but two laboratories[41, 43] using proportional counters did not observe any gamma rays around 33 kev. A source of at least a portion of the observed 33 kev radiation is thought to be a lanthanum impurity.[43, 44]

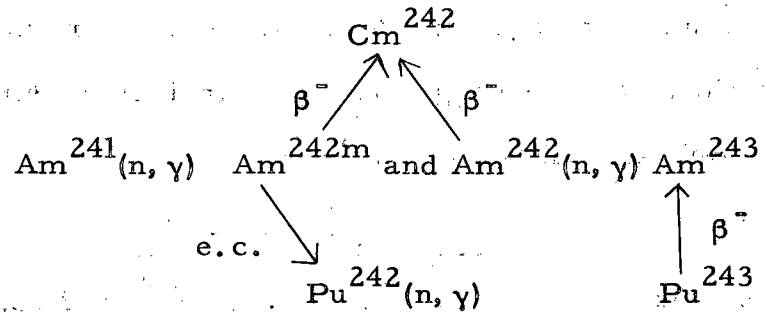
Conversion electrons corresponding to an L_{III} conversion of a 41.4 kev gamma ray of Am^{241} have been observed[35] and conversion electrons corresponding to L_I, L_{III}, M_I and N_I conversion of a 43 kev gamma in U^{237} decay[41] were ascribed to the same transition in Np^{237} as the 41 kev gamma ray in Am^{241} decay.

A gamma ray of about 100 kev was found by Martin[14] in an intensity of 10^{-4} per alpha particle.

A possible decay scheme for Am^{241} is shown in Fig. 21a. The lifetime of the 71 kev state has been reported as $6.3 \pm 0.5 \times 10^{-8}$ sec and the maximum lifetime of the 43 and 114 kev states as less than 0.3 and 1 μsec,[39] respectively. From the latter values it was deduced that all transitions from the 114 and 43 kev states were M_I, M_2, or E_2 transitions. In the case of the decay of 43 to the 11 kev level, the transition would be either M_1 or E_2 since according to Beling,[39] et al., these states would have the same parity.

F. Complex Alpha Spectrum of Am^{243}

Am^{243} can be prepared by neutron bombardment of Am^{241} from the reaction:



Previous to the present work, the measurement of alpha energy had been made with an ionization chamber coupled to a pulse height discriminator for which the value 5.27 Mev was reported.

A spectrograph sample of 2×10^7 alpha disintegrations per minute was prepared from a mixture of Am^{241} and Am^{243} activities by sublimation and exposed in the spectrograph for 87 hours. Two new alpha groups were observed which had not been seen on an exposure of pure Am^{241}.

The energy for the principal group of Am^{243} was determined by comparison with the main group of Am^{241}. If 5.476 Mev is taken as the energy of α_{71} of Am^{241}, then the main group of Am^{243} has an energy of 5.267 Mev.

A second alpha group of Am^{243} was found 41 kev lower in alpha particle energy than the most abundant alpha group and comprised about 10 percent of the total alpha particles emitted by Am^{243}.

The ratio of Am^{241} alpha activity to Am^{243} alpha activity was measured as 52. A mass analysis of the sample by F. L. Reynolds

showed that the relative masses of Am^{241}, Am^{242} and Am^{243} were 74.15, 0.615, and 25.22 percent. Thus by taking the half-life of Am^{241} as 470 years, we find a half-life of 8.3×10^3 years for Am^{243}. Our result is in good agreement with the $\sim 10^4$ year value reported by Street, Ghiorso and Seaborg from mass analysis of Am^{243} and the chemical yield of milked Np^{239}.[45]

G. Gamma Rays of Am^{243}

Previous to the present study, Martin[14] had reported a 75 kev gamma ray in Am^{243} with an abundance equal to 90 percent of the alpha transitions. In order to verify these results, alpha-gamma coincidences were run on a sample containing 700 c/m of mixed Am^{241} and Am^{243} activity. An alpha pulse analysis of the sample showed 70 percent Am^{243} activity and 30 percent Am^{241} activity. The coincidences were run between alpha particles and differential portions of the gamma spectrum. In order to calibrate the system energywise simultaneously with the coincidence measurements, a sample of Am^{241} in a closed cardboard container was placed near the gamma counter. Since none of the alpha particles in this separate Am^{241} sample could strike the ZnS screen, no true coincidences could be caused by this sample. Because of the low alpha counting rate of the mixed Am^{241} and Am^{243} sample, the chance coincidence counting rate would be quite small. From this work, after subtracting those coincidences due to Am^{241}, a 75 kev gamma ray was found to be in coincidence with 8 x 10 percent of the alpha particles. This is in excellent agreement with the previous work.

1. Decay scheme of Am^{243}. -- A decay scheme for Am^{243} is shown in Fig. 21b.

The conversion coefficient for this 75 kev gamma from the above data must be equal to or smaller than 2/8. From Gellman's tables[22] one would expect conversion coefficients of at least 5 and 15, respectively, for M_1 or E_2 radiation, while for E_1 radiation the conversion coefficient is expected to lie between 0.09 and 0.4.

That the 75 kev gamma belongs in the level scheme of Np^{239} is verified by the presence of a 73.6 kev gamma ray in the decay of U^{239}.[46] From this decay scheme, the total decay energy of Am^{243} is 5.267 + 0.088 + 0.075 Mev = 5.430 Mev. Using our energy of 6.151 Mev for the total decay energy of Cm^{243} and 0.718 Mev for the beta decay energy of Np^{239}, we find that Cm^{243} is unstable with respect to Am^{243} by 0.003 Mev. This is in good agreement with the experimentally observed beta stability of Am^{243}[45] and Cm^{243}.[47]

H. Complex Alpha Spectrum of Pu^{239}

Soon after the availability of Pu^{239} in supratracer quantities, the alpha activity was found to be accompanied by gamma radiation in low abundance.[48, 49] These and later measurements will be discussed below when the decay scheme is considered. The complex alpha spectrum presumed from these data was first observed directly by Rosenblum, Valadares and Goldschmidt[50] who employed a large permanent magnet spectrograph. They found the highest energy group in highest abundance and a prominent group at 50 kev lower energy. The present study amplifies these results and attempts to

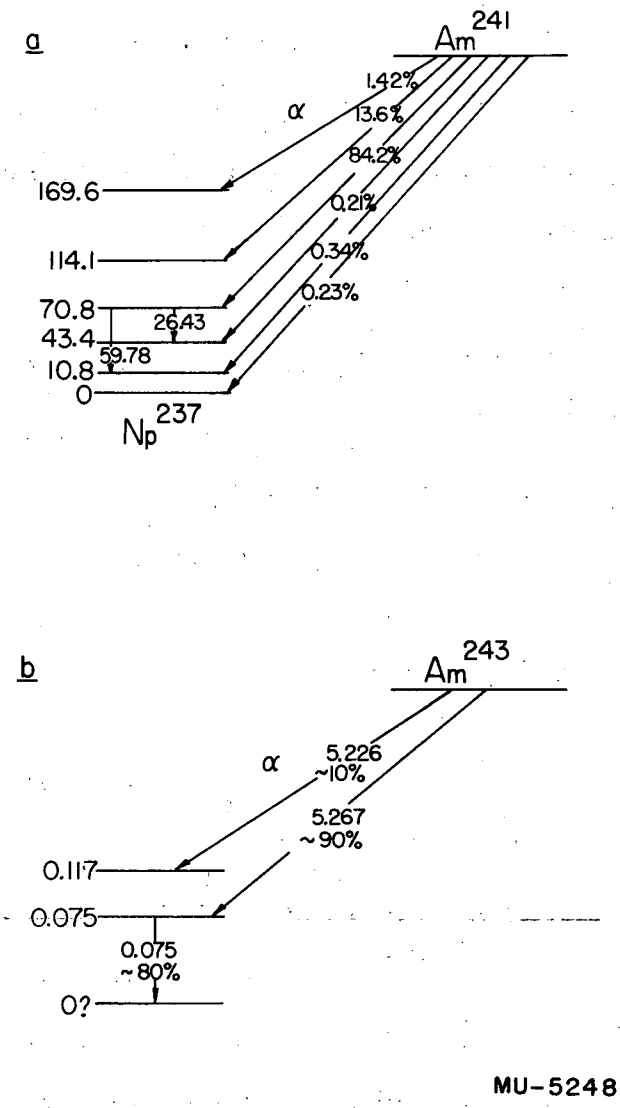


Fig. 21. a) Decay scheme of Am^{241}.

b) Decay scheme of Am^{243}.

explain the gamma ray spectrum which has been reported by several sources.

Fig. 22 shows the alpha spectrum of isotopically pure Pu^{239}.[51] The source consisted of 2.5 micrograms of Pu^{239} and was exposed for 110 hours. All three of the observed peaks have the same width at half-maximum (~ 8 kev) and the abundances as indicated were found both by comparing peak heights and by integrating the total alpha tracks.

In another exposure for 90 hours the slit system was changed to give higher transmission in order to look for rare alpha groups. The peak widths went up to 21 kev and α_o and α_{13} were no longer clearly resolved. The abundance of α_{51} was found to be 11.7 percent which agrees with the other measurements cited. A careful search was made for other alpha groups and the data are recorded in Fig. 23. The energy range covered was 4.82 - 5.57 Mev which extends from 330 kev below the main group to 430 kev above it. No alpha group was found and the limits can be set as follows: from 25 kev above α_o to 430 kev there is no group greater than 0.15 percent abundance; and from 70 kev below α_o (20 kev below α_{51}) to 330 kev there is no group in greater than 0.3 percent abundance. The plate from which Fig. 22 was derived was also counted over an extended range and the results were substantially the same although the limits of detection were not so low because of the fewer total tracks recorded. In addition, as is seen in Fig. 24 an upper limit of 0.1 percent can be set on the abundance of any alpha group from 90 kev below α_o (40 kev below α_{51}) to 190 kev below α_o. The source from which Fig. 24 was derived

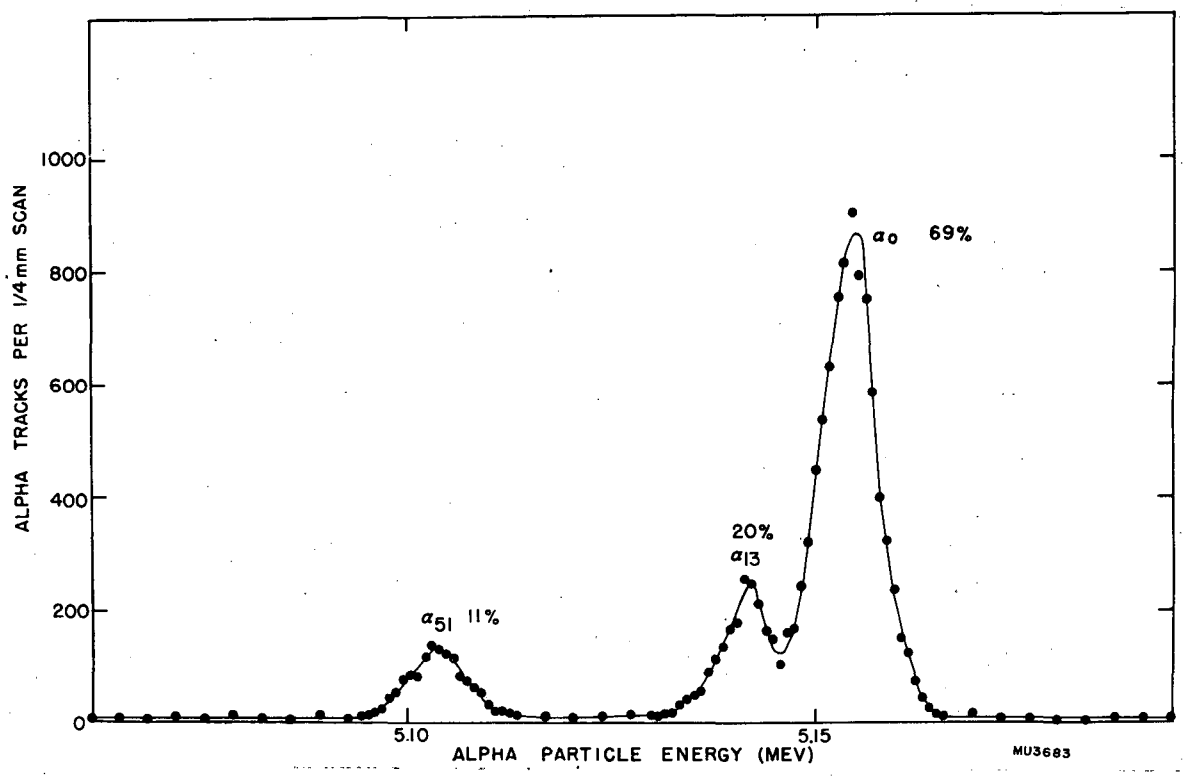


Fig. 22. Pu^{239} alpha spectrum at high resolution.

consisted of 4×10^7 alpha disintegrations per minute. A mass analysis of the plutonium from which the sample was made is shown in Table 15.

The energy for Pu^{239} α_o was determined by comparison with Po^{210} for which the alpha particle energy was taken as 5.298 Mev.[4, 5] The energy for α_o of Pu^{239} was found to be 5.150 ± 0.002 Mev as compared with the other spectrograph value,[50] 5.147, obtained using ThC as a standard.

These spectrograph values are in good agreement with reported values obtained using ionization chambers and range measurements. Cranshaw and Harvey,[52] using an ionization chamber with a pulse height analyzer, reported 5.159 ($\pm$ 0.005) Mev and Jesse and Forstat[53] obtained 5.140 ($\pm$ 0.005) Mev by measuring total ionization current. An air range determination by Chamberlain,[54] et al. gave 5.15 Mev.

The energies of the two shorter range groups were obtained from their displacements from the main group using the dispersion of the instrument as described in an earlier section. The differences in energy from the main group, α_o, were 12.8 ± 0.7 kev and 49.7 ± 0.7 kev. Rosenblum, Valadares and Goldschmidt[50] resolved α_{51} and found its energy to be 50 kev lower than the main group and our result is in excellent agreement for this group. Upon addition of a correction of 1.7 percent to these alpha particle energy differences to obtain the spacing between energy levels and rounding off to the nearest kilovolt, the level differences become 13 and 51 kev, hence α_{13} and α_{51}. These energies would correspond to gamma ray energies between the appropriate levels.

Table 15

Mass Analysis[a] of Plutonium Spectrograph Samples

Isotope	Fig. 24	Fig. 26	Fig. 27
238	1.5		
239	80.0	57.8	30.39
240	4.2	34.2	49.39
241	0.5	6.7	13.46
242	13.7	1.1	6.75

[a]These data were kindly supplied by F. L. Reynolds.

1. Decay scheme of Pu^{239}. -- It will be pointed out that the Pu^{239} alpha spectrum as observed here cannot explain all features of the gamma ray and conversion electron spectra which have been reported. First we shall examine the area of agreement which is indicated in Fig. 25.

A gamma ray of 50 kev was reported early by Ghiorso,[48] and this has been abundantly confirmed. Freedman, et al.[35] found conversion electrons of a 53 kev gamma besides observing the gamma ray with a proportional counter and a scintillation spectrometer. West and Dawson,[55, 56] using a proportional counter spectrometer, found a 52.0 ± 0.3 kev photon in an abundance of 7×10^{-5} per alpha particle. Martin[14] in this laboratory has measured this gamma ray as 53 ± 2 kev with a scintillation spectrometer, while two laboratories have found conversion electrons corresponding to a 50 kev gamma ray in coincidence with alpha tracks in Pu^{239} impregnated emulsions. Albouy and Teillac[57] found the electrons to be present to the extent of 16 per 100 total alpha particles while Dunlavey and Seaborg[16] found 12.5 electrons per 100 alpha particles. It seems almost certain that this gamma ray represents the transition between the levels reached by α_{51} and α_0 because of the agreement in energy and moderate agreement in abundance. Rosenblum and co-workers[50] reported a much higher abundance than ours for α_{51}, but their resolution would have been more difficult.

West and Dawson[56] also found a gamma ray of 38.5 ± 0.5 kev in an abundance of 2/7 of their 52.0 kev gamma. Freedman, et al.[35] similarly reported a 39 kev gamma ray in an abundance

of 2/7 of their 53 kev gamma. In addition, some of the conversion lines attributed by Freedman, et al.[35] to L_I and L_{II} conversion of a 53 kev gamma could be due, at least in part, to M conversion of their 39 kev gamma.

The preceding discussion shows that there need be no sharp disagreement between our alpha spectrum and the gamma ray transitions so far considered. However, there is harder gamma radiation which is more difficult to explain. There have been reported gamma rays of 384 kev,[35] 300 kev,[48] and 420 kev[49] in very low abundance and these could well correspond to low energy alpha groups in quantity below our level of detection. In addition, Freedman, et al.[35] found gamma rays of 100 and 124 kev, and another gamma ray can be deduced from the work by Albouy and Teillac[57] and by Dunlavey and Seaborg[16, 58] who both measured electron tracks stemming from alpha tracks in nuclear emulsions. Albouy and Teillac reported theirs as the "K conversion electrons from a 200 kev gamma ray" which would mean that the electron line was 100 kev and the abundance as 0.1-1 percent of the alpha particles. It is doubtful that these electrons are K shell converted because the corresponding K x-rays have not been seen in the requisite abundance. West, et al.[59] report 101 and 115 kev K x-rays in an intensity of 2×10^{-5} per alpha particle. Dunlavey and Seaborg found similar electrons in the energy range 100 ± 20 kev in 0.5 percent abundance.

It seems likely that these electrons are L conversion lines of ~120 kev gamma rays corresponding to the 124 kev gamma found

by Freedman, et al.[35] If the abundance of the electron is correct, the corresponding alpha group should have been seen. According to the data of Fig. 23, an alpha group of lower energy than α_0 by 70 kev or more would be detected if present in 0.3 percent abundance or greater. A still lower limit was obtained according to the data in Fig. 24. From 90 kev below α_0 to 190 kev there is no alpha group greater than 0.1 percent. The discrepancy between the abundance of the 120 kev transition and the absence of a corresponding alpha group is large enough to cause some doubt as to the ground state transition of Pu^{239}.

A possible decay scheme is shown in Fig. 25. The U^{235} level populated by the most abundant alpha group of Pu^{239} would be a metastable state. A search is being made for any long-lived radiation which might occur from this level.

I. Complex Alpha Spectra of Pu^{240} and Pu^{241}

This isotope of plutonium is an alpha emitter of 6600-year half-life[60] and is best prepared by neutron capture by Pu^{239}. Its alpha energy is known to be very close to that for Pu^{239} and had not been previously resolved for sure.

A sample containing 5×10^5 disintegrations per minute of mixed Pu^{239} and Pu^{240} activities was exposed for 46 hours, and the spectrum shown in Fig. 26 was registered. The peaks were readily assigned to their respective isotopes because the mass spectrographic analysis shown in Table 15 indicated that Pu^{240} was in abundance such that its alpha activity should predominate and the energy differences and relative abundances of the three peaks assigned to Pu^{239} were in close agreement with those for Pu^{239} (see Fig. 22).

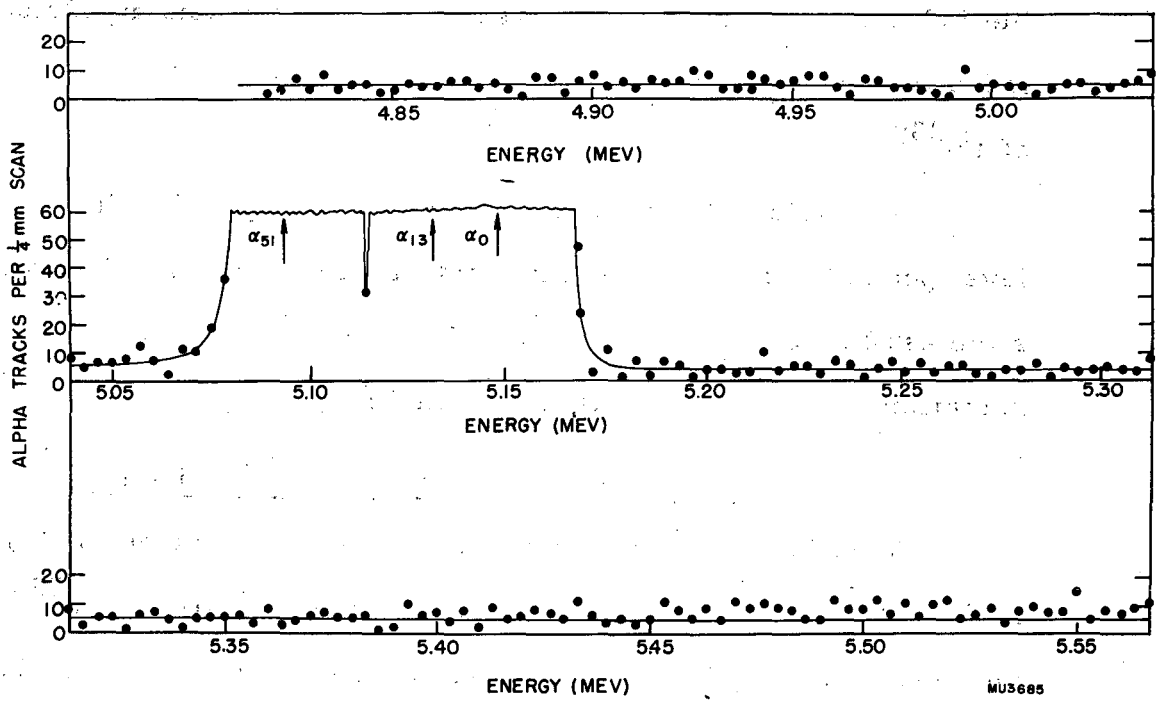


Fig. 23. Pu^{239} alpha spectrum showing extended energy regions.

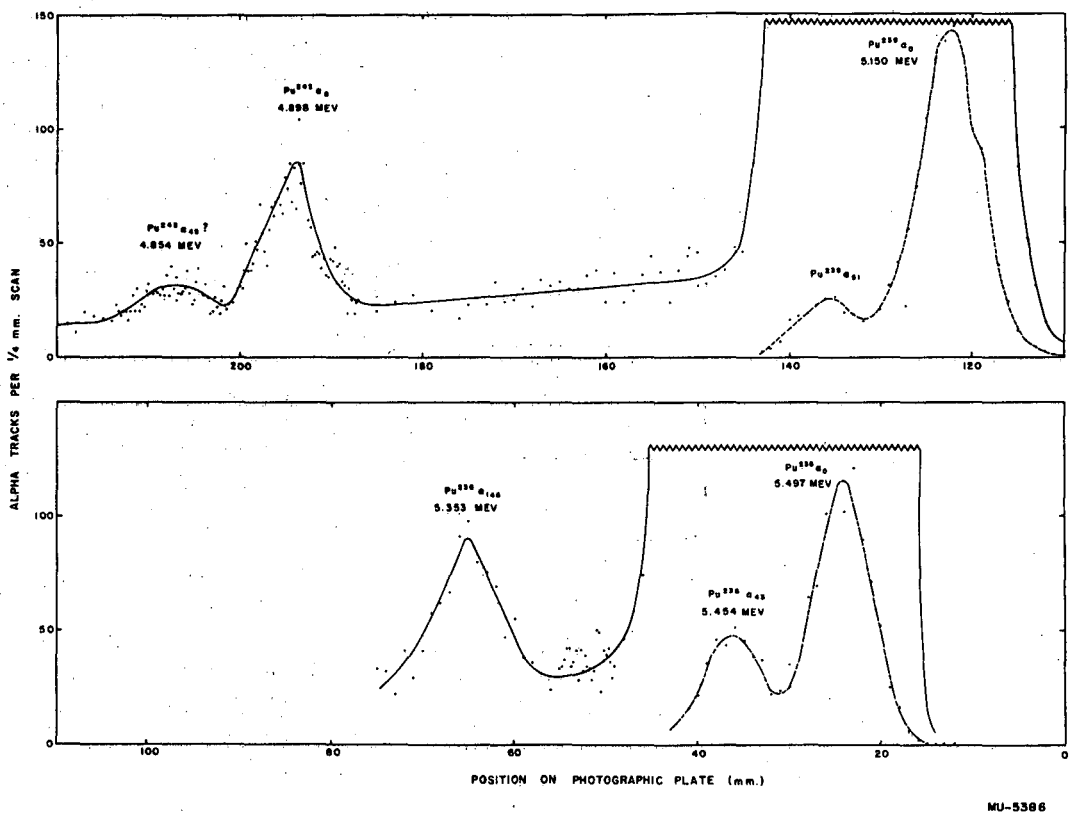


Fig. 24. $Pu^{238, 239, 242}$ alpha spectra.

——— Full microscope scan.

-- -- -- About 1/70 full scan.

— - — About 1/300 full scan.

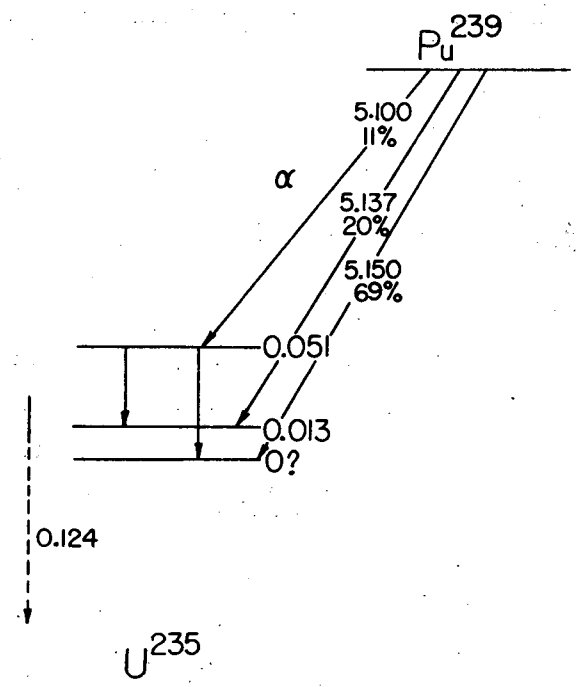


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Fig. 25. Pu^{239} decay scheme.

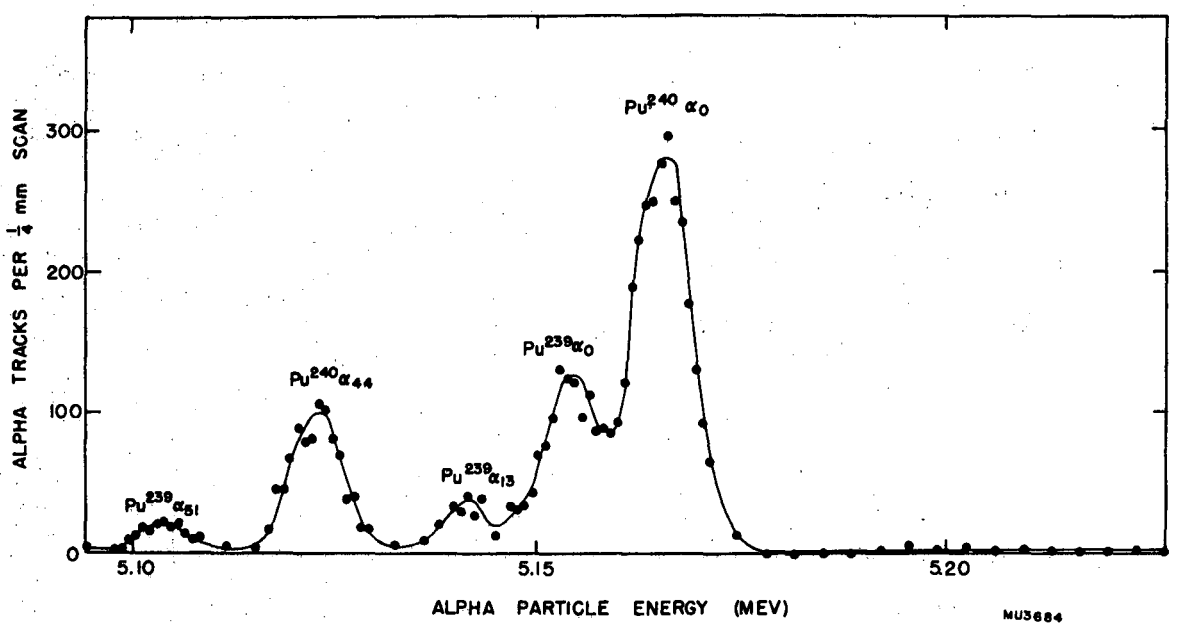

Fig. 26. $Pu^{239,240}$ alpha spectra.

The energy of the main group of Pu^{240} is 12 $\pm$ 2 kev greater than that for Pu^{239}. Using our value, 5.150 Mev for the energy of the α_0 group of Pu^{239}, the energy for Pu^{240} α_0 group is 5.162 Mev. Since we are fairly certain that the most abundant group of an even-even alpha emitter represents the ground state transition, we may then calculate the decay energy for Pu^{240} as 5.250 Mev.

The second alpha group of Pu^{240} is 43.5 $\pm$ 2 kev lower in energy than the main group; hence it is designated α_{44} signifying that it leads to a state 44 kev above the ground state. Its abundance is 24 percent, therefore the partial alpha half-lives for the two groups, based on the 6600 year measured half-life are 8700 years and 27,000 years.

The isotope Pu^{241} is generally made by neutron irradiation of Pu^{239} by the reaction:

$$Pu^{239}(n, \gamma)Pu^{240}(n, \gamma)Pu^{241}.$$

It is a beta emitter with a 14-year half-life.[61] Work done previous to this study indicated a partial alpha half-life of about 4×10^5 years and an alpha particle energy of 4.91 Mev as measured with an ion chamber.[61]

A sample of 10^7 alpha disintegrations per minute of mixed $Pu^{239, 240, 241, 242}$ was exposed for 88 hours and the spectrum shown in Fig. 27 was registered. The mass analysis is given in Table 15. The peaks all have a half-thickness of about 7 mm or 24 kev. This comparatively large half-thickness is due to the large slit opening used at the source, 1/8 in. x 1 in.

The two intense alpha groups shown in Fig. 27 are separated by 45 kev and are due to Pu^{240} with a small contribution from Pu^{239} as deduced from the mass analysis. The particle energies of all groups shown in Fig. 27 were determined using 5.162 Mev as the energy of Pu^{240} α_o.

The three remaining alpha groups could be due to Pu^{240} or Pu^{241}. Pu^{242} is ruled out because of half-life considerations. In order to establish a definite assignment to either Pu^{240} or Pu^{241} another exposure will be made with a very active sample with isotopic composition as shown in Fig. 26. Until this experiment is completed, the alpha groups at 4.893 and 4.848 Mev are tentatively assigned to Pu^{241} and the alpha group at 5.014 Mev is assigned to Pu^{240}. This assignment to Pu^{240} is based on the similarity of the Pu^{240} alpha spectrum to the Cm^{242} alpha spectrum. By a comparison of peak heights the alpha group at 5.014 Mev is 0.1 percent of the total Pu^{240} alpha particles. Since this group is 151 kev lower in decay energy than the main group of Pu^{240}, it is designated α_{151}.

The two alpha groups assigned to Pu^{241} are separated by 46 kev in decay energy. The group at 4.848 Mev comprises about 25 percent of the groups assigned to Pu^{241}.

The Pu^{241} alpha branching ratio may be determined from the ratio of the Pu^{240} and Pu^{241} masses, their alpha abundances and their half-lives. From this data an alpha half-life of 3×10^5 years is in excellent agreement with the previously reported value of 4×10^5 years.[61]

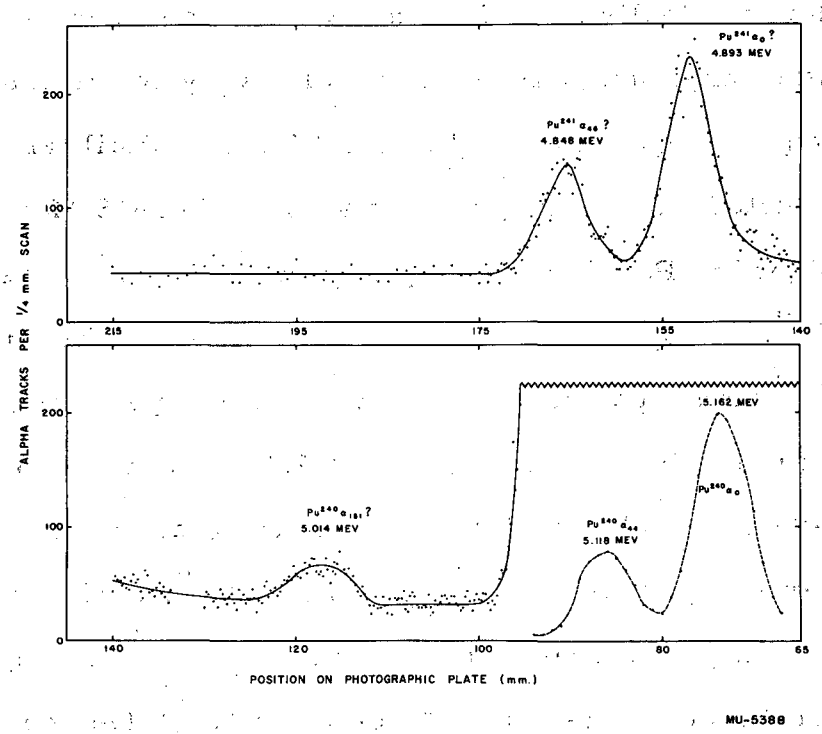

Fig. 27. $Pu^{240, 241}$ alpha spectra.

——— Full scan.

— — — About 1/140 full scan.

1. Decay schemes of Pu^{240} and Pu^{241}. -- Freedman, et al.[35] reported conversion lines of a 49.6 kev gamma ray and measured the energy of the gamma quantum with a NaI crystal spectrometer and a proportional counter as 47 and 48 kev. West[59] reported a 45.0 ± 0.2 kev gamma ray present in the alpha decay of either Pu^{240} or Pu^{238}. These transitions may correspond to the transition from the excited state in U^{236} populated by Pu^{240} α_{44} to the ground state of U^{236} as shown in Fig. 28a. From the ratio of L x-rays to gamma quanta determined by Freedman, et al., a lower limit of 35 can be set on the conversion coefficient of this transition. The actual value of the conversion coefficient would depend on the L Auger coefficient and the extent of the absorption of L x-rays before they enter the active body of the NaI crystal. No radiation has been observed from the level populated by Pu^{240} α_{151}.

Freedman, et al.[35] report 100 kev and 145 kev gamma rays in Pu^{241} with abundances, respectively, of 0.35 and 0.07 per alpha particle. Because of its high abundance, the 100 kev radiation is probably in coincidence with the alpha group at 4.893 Mev. Freedman reported that the half-width of the 100 kev radiation appeared to be too large for a single gamma ray, and that this width was very nearly equal to that expected for uranium x-rays. If the 100 kev transitions are K x-rays due to K conversion of the 145 kev gamma ray, the K conversion coefficient would be about 35/7 = 5 (the K Auger coefficient is assumed to be small). This value of the conversion coefficient may be interpreted from Rose's[21] tables as corresponding to an M_3 transition.

A possible decay scheme is shown in Fig. 28b.

J. Complex Alpha Spectrum of Pu^{242}

Pu^{242} is generally made by the electron capture decay of Am^{242} by the reaction:

$$Am^{241}(n, \gamma)\ Am^{242}\ (16\ hr)\ Am^{242}\ (100\ y)$$
$$\downarrow \text{e.c.}$$
$$Pu^{242}$$

It is a beta stable alpha emitter with a particle energy of 4.88 Mev[61] as determined with an ion chamber previous to this study. A sample of 3.7×10^7 plutonium alpha dis/min was exposed for 37 hours. This is the sample whose alpha spectrum is shown in Fig. 24 and whose mass analysis is shown in Table 15.

The energy of the alpha group at 4.898 Mev was determined by comparison with the Pu^{239} α_o peak on the same plate. The Pu^{239} α_o peak would be affected by the Pu^{240} α_o peak and the Pu^{239} α_{13} peak. These effects, however, tend to cancel each other out, so the energy of the Pu^{239} α_o was taken as 5.150 Mev. The 4.898 Mev alpha group could not be due to Pu^{239} as it would have been seen in Fig. 23. Likewise it could not be due to Pu^{240} as it would have been seen in Fig. 27 in about four times the abundance of any group in that region. If the group were due to Pu^{241} it should have been seen in Fig. 27 in about ten times the abundance of any group observed in that region. The alpha group shown at 4.898 Mev, therefore, must belong to Pu^{242}.

A second alpha group 44 kev lower in particle energy than Pu^{242} α_o was found with an abundance of about 20 percent of all the groups assigned to Pu^{242}. By similar reasoning to that used above, this alpha group cannot belong to either Pu^{240} or Pu^{241}. It could, however, be a low energy alpha group of Pu^{239} or Pu^{242}. Pending

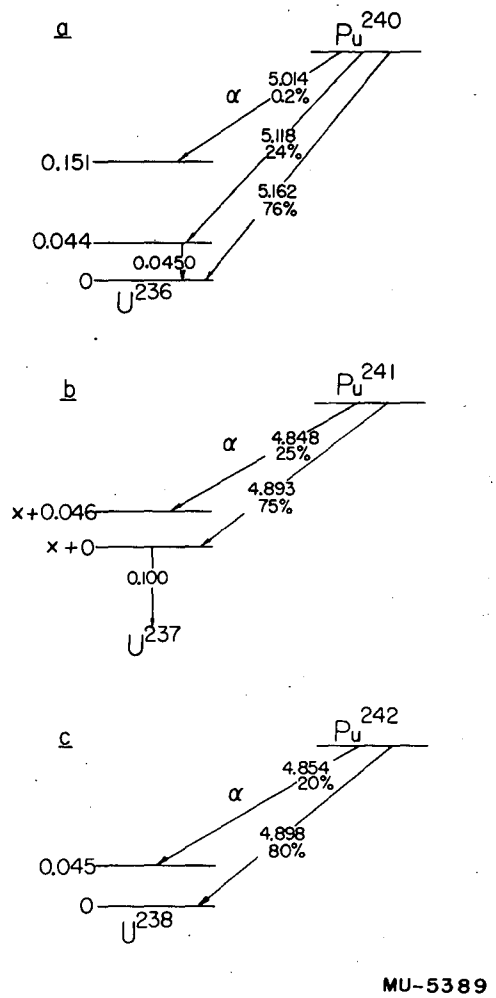

Fig. 28. a) Pu^{240} decay scheme.

b) Pu^{241} decay scheme.

c) Pu^{242} decay scheme.

further experiments on pure Pu^{239}, the alpha group at 4.854 Mev is tentatively assigned to Pu^{242}. This assignment to Pu^{242} is made because of the similarity to the alpha decay characteristics of other even-even nuclides as discussed in a later section.

The half-life of Pu^{242} can be determined from the relative atomic masses and alpha activity of Pu^{239} and Pu^{242} in the sample. Using a half-life of 24,360 years for Pu^{239}, we find the half-life of Pu^{242} to be 9×10^5 years. This compares with the value of 5×10^5 years[61] found by Thompson, et al.

A decay scheme for Pu^{242} is shown in Fig. 28c.

K. Complex Alpha Spectrum of Pu^{238}

Pu^{238} is most easily prepared by milking its parent, Cm^{242}. It has a half-life of 89.6 years.[62] Previous to this study the alpha particle energy had been determined by the range of the particles in air giving values of 5.47[63] and 5.51 Mev.[54]

A sample of 7×10^7 alpha disintegrations per minute of Pu^{238} was exposed in the spectrograph for 114 hours. Its spectrum is not shown. The sample mentioned before in connection with Pu^{242} (see Fig. 24) contained a large amount of Pu^{238} alpha activity. The spectrum of Pu^{238} shown in Fig. 24 was in good agreement with respect to energy separation and relative abundances with the Pu^{238} spectrum which is not shown.

Since the masses of Pu^{239}, Pu^{240}, and Pu^{242} varied by over an order of magnitude in the two samples relative to Pu^{238}, the assignment of the groups to Pu^{238} is positive.

The most energetic alpha group has an abundance of 76 percent of the total Pu^{238} alpha emission and will be designated α_0 of Pu^{238}.

The alpha particle energy of α_o was determined by comparison with the main alpha group of Pu^{239} as shown in Fig. 24. The energy of the Pu^{239} group was taken as 5.150 Mev giving an energy of 5.497 Mev for Pu^{238}.

Another energy determination was made by exposing an old sample of 10^8 alpha disintegrations per minute of Cm^{242}. The two intense Pu^{238} peaks were easily detectable, and the energy of the Pu^{238} α_o was measured as 5.492 Mev. The energy of Cm^{242} α_o was taken as 6.110 Mev. With an averaged particle energy of 5.495 Mev, the resulting decay energy of Pu^{238} is 5.589 Mev.

A second alpha group 42 kev lower in particle energy than Pu^{238} α_o has an abundance of 24 percent of the total Pu^{238} alpha emission. Adding a correction of 0.7 kev to the particle energy separation and rounding off to the nearest kilovolt, we find an energy of 43 kev for the corresponding state in the residual nucleus. This alpha group is designated α_{43} of Pu^{238}.

The third alpha group is 144 kev lower in particle energy than the main group and has an abundance of 0.15 percent of the total Pu^{238} alpha emission from peak height comparisons. Adding a correction for nuclear recoil, the decay energy separation becomes 146 kev and the alpha group is designated Pu^{238} α_{146}.

L. Gamma Rays of Pu^{238}

The sample of Pu^{238} whose alpha spectrum is not shown was gamma analyzed, and gamma rays of 17, 44, 101 and 149 kev were observed as shown in Fig. 29. The respective relative gamma intensities for the 44, 101 and 149 kev radiations were 100, 27 and 3.3.

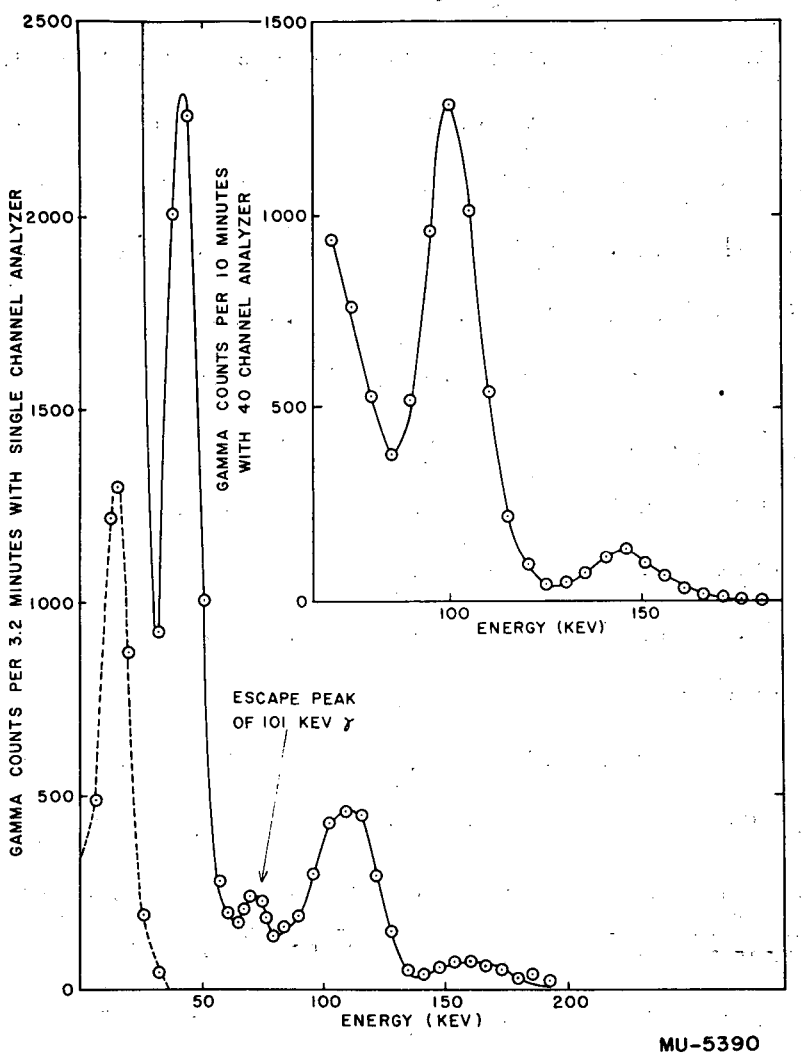

Fig. 29. Gamma rays of Pu^{238}.

No correction was made for changes in crystal counting efficiency with energy. The effect is expected to be small. The abundance of the 44 kev gamma quanta was measured as 3.8×10^{-4} per alpha particle. The geometry of the alpha analyzing system was determined with Am^{241} by assuming the 60 kev gamma ray had an abundance of 40 percent[6] per Am^{241} alpha particle.

The energy of the 44 kev gamma was the average of two values, 42 and 45 kev, and the abundance per alpha particle was the result of one measurement.

The energy of the 101 kev gamma was the average of five values which ranged from 96 to 108 kev. The abundance of this gamma ray with respect to the 44 kev gamma was the average of two measurements, 24 percent and 30 percent.

The energy of the 149 kev gamma was the average of four measurements ranging from 145 to 157 kev. The abundance with respect to the 101 kev gamma (12 percent) was the average of three measurements which ranged from 12 to 13 percent.

The 17 kev radiation is probably due to L x-rays. Radiation of this energy is so strongly absorbed in the counting system that no accurate intensity can be determined.

1. Decay scheme of Pu^{238}. -- Prior to this study, Reed[64] found L x-rays, a 42.6 kev gamma, and a possible 166 kev gamma by absorption measurements. O'Kelley[15] found electrons corresponding to a 45 kev gamma ray and a possible 48 kev gamma ray. Coincident with this study, Dunlavey and Seaborg[16] found conversion electrons corresponding to a 40 kev gamma ray in

coincidence with 23 ± 3 percent of the alpha particles.

The 44 kev gamma ray observed in this study and by other workers probably corresponds to the transition between the excited state in U^{234} populated by α_{43} of Pu^{238} and the ground state.

The conversion coefficient of this transition from our data would be:

$$\frac{0.24 - 0.00038}{3.8 \times 10^{-4}} = 6.4 \times 10^{2}.$$

If the same M/L ratio is taken as that used for Cm^{242},[16] the L conversion coefficient would be 5.3×10^{2}. From Gellman's tables[22] one would expect conversion coefficients of 1, 6 x 10, and 6×10^{2}, respectively, for E_1, M_1 and E_2 transitions. Because of the excellent agreement with the E_2 transition, a likely spin assignment for the first excited state is 2 even parity.

The observed 101 kev gamma is in good agreement with the transition from the level in U^{234} populated by α_{146} to the level populated by α_{43}. Assuming that all of this gamma ray is caused by the transition between these two levels, and that no other decay takes place from the 146 kev level, we find a conversion coefficient of:

$$\frac{(1.5 - 0.1) \times 10^{-3}}{1.0 \times 10^{-4}} = 14.$$

The ratio of M to L conversion is not known, but it should not be larger than for the Cm^{242} 44 kev transition.[16] Therefore, the L conversion coefficient of the 101 kev gamma would be at least 11. From Gellman's tables,[22] the theoretical conversion coefficients for E_1, M_1 and E_2 radiation are 0.1, 4 → 9, and 6 → 26, respectively.

The exact shape of the theoretical curve is not known since Gellman's data includes only three points, and this accounts for the spread in the theoretical values. The 101 kev gamma ray could correspond to either an M_1 or an E_2 transition.

If the 101 kev radiations were K x-rays of the 149 kev gamma ray, the K conversion coefficient would be 27/3.3 = 8. From Rose's table[21] of theoretical K conversion coefficients, this would correspond to an M_1 transition or an electric transition >5. The latter case is obviously an impossibility from lifetime considerations. The M_1 transition could not represent a transition from the level in U^{234} populated by α_{146} of Pu^{238} since, as stated previously, all transitions in even-even nuclei to the ground state from levels populated by alpha decay must be electric transitions.

There is not sufficient experimental evidence to determine with certainty the place of the 101 kev radiation (or the 149 kev radiation) in the Pu^{238} decay scheme. Because of empirical correlations to be discussed in a later section, we prefer the assignment of the bulk of the 101 kev radiation to the transition from the second excited state to the first excited state of U^{234} as shown in Fig. 30.

M. Complex Alpha Spectrum of U^{234}

U^{234} can be obtained from natural uranium ores. Before a sample suitable for the spectrograph can be obtained, the U^{234} must be separated from the relatively large amount of U^{238} mass. After a quantity of U^{234} free from U^{238} was obtained, a sample of about 8 μg was prepared for the spectrograph by vacuum sublimation and

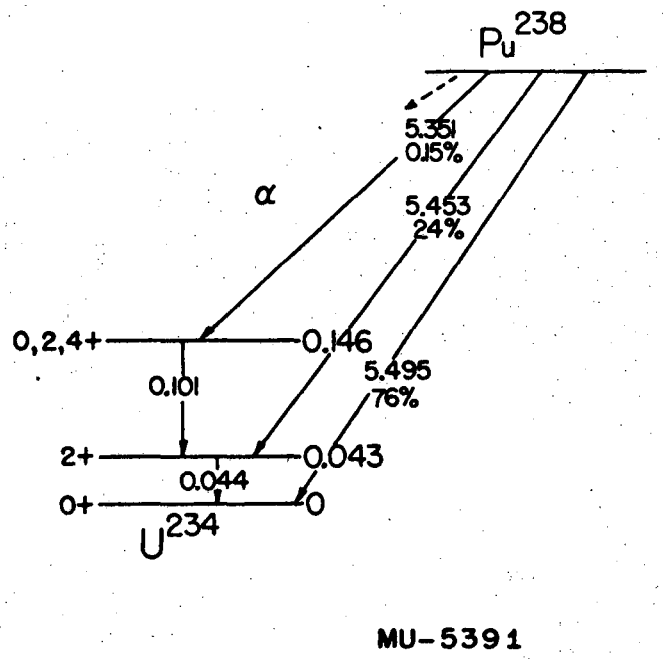

Fig. 30. Decay scheme of Pu^{238}.

and was exposed in the spectrograph for 19 hours.

Two alpha groups were found separated by 49 kev in particle energy. An earlier measurement of this separation, determined by extrapolating high energy edges, gave a value of 46 kev.

The most energetic group, designated α_0, comprised 74 percent of the observed U^{234} alpha emission. The second group, designated α_{50}, comprised 26 percent of the observed U^{234} alpha emission.

N. Gamma Rays of U^{234}

The gamma spectrum of a large sample of U^{234} was measured with the single-channel gamma analyzer and radiations of 18, 53, 90 and 120 kev were observed. The uncorrected relative intensities of the 53, 90 and 120 kev radiations were 4.5:1:2.1, respectively.

The escape peak of the 120 kev radiation would be at 87 kev and would comprise a part or all of the 90 kev radiation. The ratio of the intensities of the 120 kev gamma and its escape peak was assumed to be the same as for the Am^{241} 60 kev gamma ray and its escape peak. The resulting relative intensities after subtraction of escape peaks for the 53, 90 and 120 kev radiations were 7:1:3.5.

Unfortunately, there was a considerable amount of absorber between the U^{234} and the NaI detector. If one assumes that the relatively small amount of plutonium L x-rays observed (18 percent of 53 kev gamma ray) is due to absorption of the L x-rays before striking the NaI crystal, the absorber is equivalent to nearly 2 g of aluminum. Upon correction of all the intensities on the basis of this quantity of aluminum absorber, the relative intensities of the 53, 90 and 120 kev radiations become 15:1:3.4.

Macklin and Knight[65] observed with a Geiger counter L x-rays of thorium in the alpha decay of U^{234}. Scharff-Goldhaber and McKeoun[66] verified these results with a proportional counter. Using a more intense source, Scharff-Goldhaber[67] found gamma rays of 50 and 117 kev with a NaI scintillation counter. Bell, et al.[68] found gamma rays of 17 ± 2, 53 ± 2, 93 ± 2.5, and 118 ± 2 kev with a scintillation counter. The relative intensities of the 53, 93 and 118 kev radiation were 1.0, 0.11-0.31 and 0.42.

The gamma rays observed by other workers are in excellent agreement energywise with those of this study. The relative intensities do not agree very well, but this measurement is more difficult than the energy determinations. If the 90 kev radiation represents the K conversion of the 120 kev gamma ray, the K conversion coefficient would be 0.3 from this study and 0.26 → 0.74 from the work of Bell, et al. From Rose's tables[21] of theoretical K conversion coefficients, the values for E_1, E_2, E_3 and E_4 multipole orders are 0.2, 0.4, 0.7 and 2, respectively. For an M_1 transition the K conversion coefficient would be nearly 10. Therefore, the 120 kev gamma must represent an E_1, E_2 or E_3 transition regardless of the source of the 90 kev radiation.

L conversion of the 120 kev gamma should give electrons of about 100 kev energy. The possibility exists that these electrons may correspond to those found by Teillac[68] and reported as about "the order of 75 kev," in an intensity of ~1 percent of the U^{234} alpha emission.

Because of empirical correlations discussed in a later section, the 120 kev gamma ray is assumed to be in cascade with the

gamma rays of about 50 kev. Teillac[68] found electrons of 36 and 50 kev corresponding to L and M conversion of a 55 kev gamma ray in coincidence with 32 percent of the alpha particles.

Dunlavey and Seaborg[16] reported electrons of 30 and 45 kev corresponding to L and M conversion of a 50 kev gamma ray in coincidence with 25 percent of the alpha particles. Prohaska[38] found 40 kev electrons in coincidence with 17 percent of the alpha particles.

1. Decay scheme of U^{234}. -- The gamma rays of about 50 kev probably correspond to the transition between the two levels in Th^{230} populated by alpha decay. A possible decay scheme is shown in Fig. 32a.

O. Complex Alpha Spectrum of U^{233}

U^{233} can be obtained from neutron bombardment of Th^{232} followed by two successive beta decays. A sample of U^{233} adequate for the spectrograph was prepared by sublimation and exposed for 94 hours. The spectrum is shown in Fig. 31. Three alpha groups were observed with abundances if 83, 15 and 2 percent. The group comprising 83 percent was the most energetic and is designated α_o. The group of 15 percent abundance is 42 kev lower in decay energy than α_o and is designated α_{42}. The group of 2 percent is 96 kev lower in decay energy than α_o and is designated α_{96}.

1. Decay scheme of U^{233}. -- West, et al.[59] report L x-rays of thorium and gamma rays of 42.8 ± 0.3 and 56.1 ± 0.4 kev in intensities of $(4 \pm 1) \times 10^{-2}$, 5×10^{-4}, and 1×10^{-4} per alpha particle, and Bisgård[69] found conversion electrons corresponding to

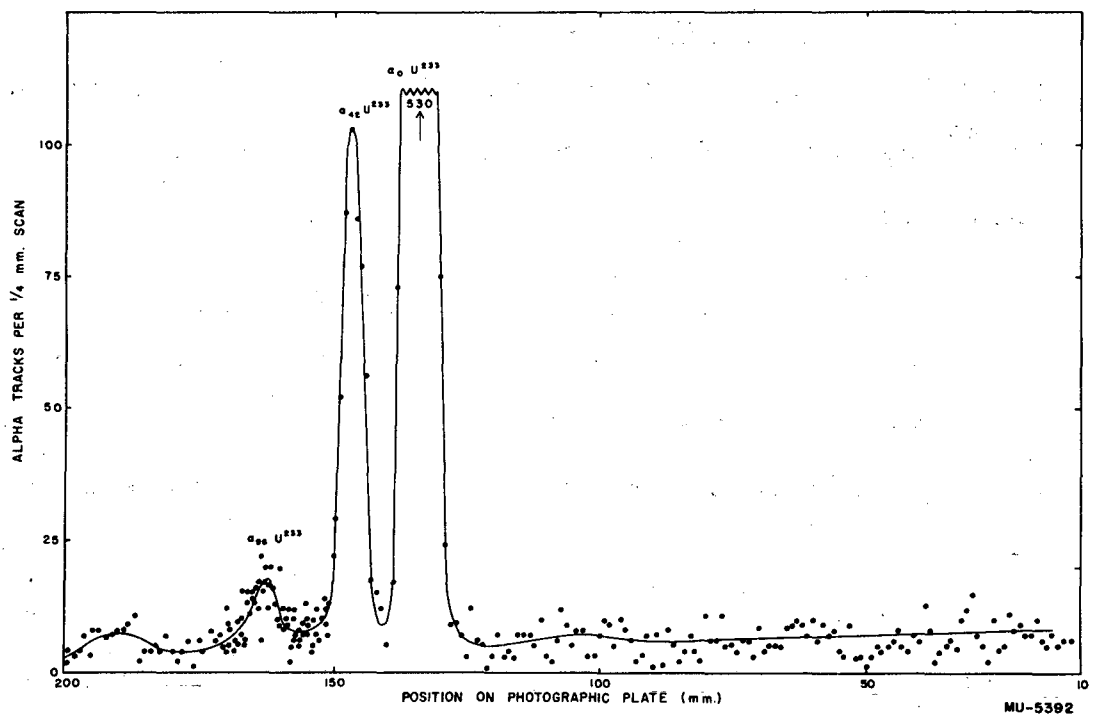

Fig. 31. U^{233} alpha spectrum.

——— 14 μgm sample.

— — — 0.3 μgm sample. Ordinate is 200 times full scan.

gamma rays of about 43 and 56 kev in coincidence with U^{233} alpha particles. Dunlavey[58] found electrons corresponding to a 40 kev gamma in coincidence with 9 percent of the alpha particles and Studier[70] observed a gamma ray of 40 kev and conversion electrons corresponding to this gamma ray. Martin[14] observed a 39 kev gamma ray in an intensity of 1 percent of the L x-rays. Prohaska[38] found a gamma ray of 39 kev in coincidence with 0.2 percent of the alpha particles. The 43 kev transition probably takes place between the level populated by α_{42} and α_o and the 56 kev transition probably takes place between the levels populated by α_{96} and α_{42}.

Studier[70] found gamma rays of 80 kev and corresponding conversion electrons. Martin[14] found an 85 kev radiation in an intensity of 1 percent of the L x-rays. Dunlavey[58] found electrons corresponding to a 90 kev gamma in coincidence with 0.4 percent of the alpha particles and Bisgård[69] found electrons corresponding to a 99 kev gamma in coincidence with the alpha particles. Prohaska[38] found a gamma ray of 88 kev. This 80-100 kev radiation may correspond to the transition between the levels populated by α_{96} and α_o.

In addition, Studier[70] found a 0.31 Mev gamma ray in an abundance of 0.1 percent. The decay scheme is shown in Fig. 32b.

P. Complex Alpha Spectrum of U^{232}

U^{232} can be prepared by neutron bombardment of Th^{230} by the reaction:

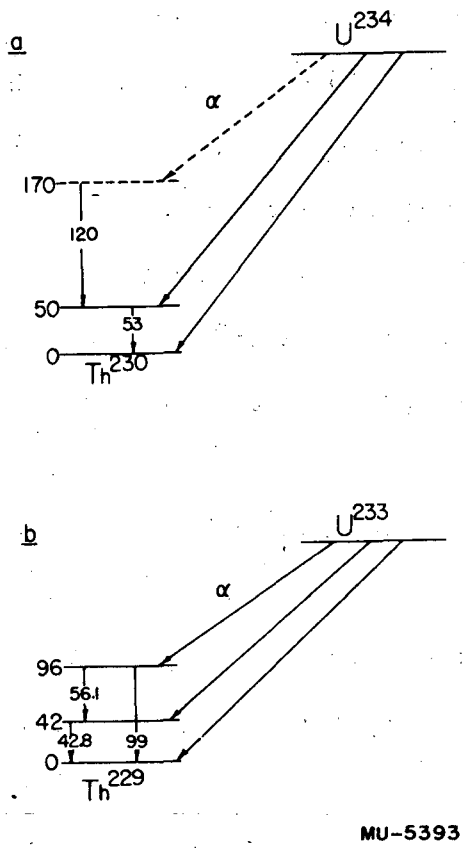

Fig. 32. a) U^{234} decay scheme.

b) U^{233} decay scheme.

$$U^{232} \xleftarrow{\beta^-} Pa^{232}$$

$$Pa^{231}(n, \gamma)\, Pa^{232}$$

$$Pa^{231} \xleftarrow{\beta^-} Th^{231}$$

$$Th^{230}(n, \gamma)\, Th^{231}$$

A sample of 10^5 alpha disintegrations per minute of U^{232} was prepared by vacuum sublimation and exposed for 46 hours. Two alpha groups separated by 57 kev in alpha particle energy were observed. The most energetic group had an abundance of 69 percent of the total U^{232} alpha particles and was designated α_o. The other alpha group was 58 kev lower in decay energy and was designated α_{58}. It constituted 31 percent of the U^{232} alpha emission.

Another sample of 3×10^8 c/m of U^{232} was prepared by vacuum sublimation and exposed in the spectrograph for 14 hours.

An additional alpha group was observed 189 kev lower in decay energy than α_o and in an abundance of 0.3 percent of the total U^{232} alpha emission. It is designated α_{189}.

The alpha particle energy of U^{232} was determined by comparison with its Th^{228} daughter. The energy of α_o of U^{232} was 5.318 Mev. Only one alpha group of Th^{228} was observed. We assumed this group was α_o of Th^{228}.

Q. Complex Alpha Spectra of U^{230}, Th^{226} and Ra^{222}

U^{230} can be prepared by the reaction:

$$\begin{array}{c} U^{230} \\ \uparrow \beta^- \\ Th^{232}(p, 3n)\ Pa^{230} \end{array}$$

Th^{226} and Ra^{222} are formed by succeeding alpha decay processes of U^{230}.

A sublimed sample of U^{230} was exposed in the spectrograph for 111 hours and the resulting spectrum is shown in Fig. 33. The most intense alpha groups in nearly equivalent abundance were identified by comparing their energy separations with those determined by other workers from ion chamber measurements. The smaller intensity groups were assumed to belong to the isotope whose main group was nearest in energy on the high energy side.

U^{230} has two alpha groups separated by 71 kev in decay energy. The most energetic group comprises 77 percent of the total U^{230} alpha emission and is designated α_0 of U^{230}. The lower energy group comprising 23 percent* of the U^{230} alpha emission is designated α_{71}. An earlier measurement was made by comparing the differences between the extrapolated edges, and the resulting alpha decay separation between the U^{230} groups was given as 70 kev.

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*Later results indicate this value may be ~35 percent. The listed measurement was made with U^{230} α_{71} at the extreme edge and partially off of the photographic plate.

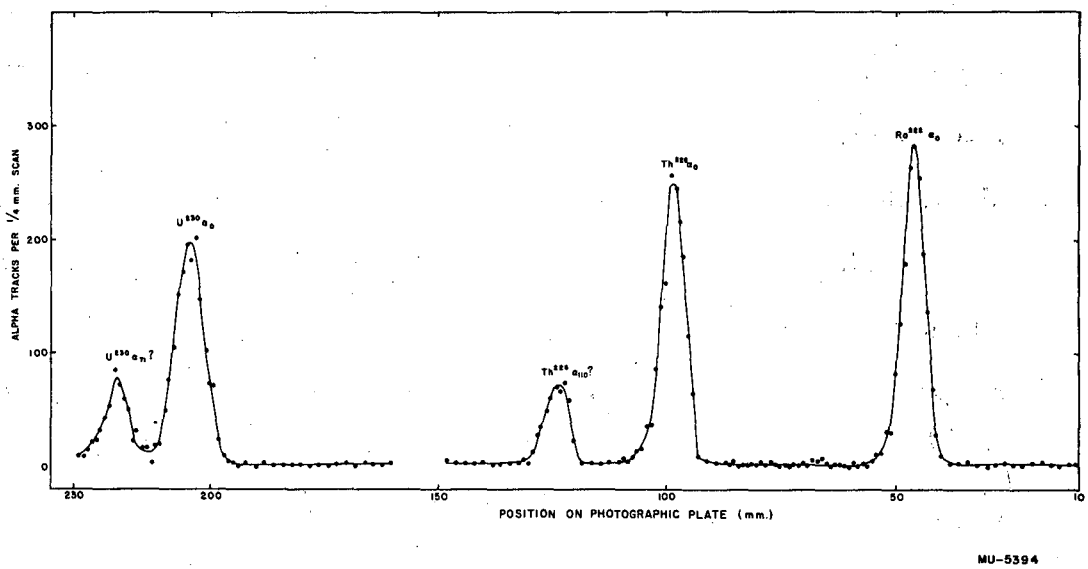

Fig. 33. U^{230}, Th^{226}, Ra^{222} alpha spectra.

The main group of Th^{226} was 456 kev higher in particle energy than α_o of U^{230} and comprises 78 percent of the total Th^{226} alpha emission. This group is designated α_o of Th^{226}. A second group 110 kev lower in decay energy than α_o comprises 22 percent of the total alpha emission and is designated α_{110} of Th^{226}. An earlier measurement using the difference in extrapolated high energy edges resulted in a difference of 117 kev between the decay energies of the Th^{226} alpha groups.

The only observed alpha group of Ra^{222} was found 681 kev higher in energy than α_o of U^{230}. From correlations made in a later section, the second alpha group of Ra^{222} is expected to be at about the same energy as α_{110} of Th^{226} but in much lower intensity.

The energies of U^{230}, Th^{226} and Ra^{224} were measured by another worker[71] with an ion chamber as 5.85, 6.30 and 6.51 Mev, respectively. The energy separations of Th^{226} and Ra^{222} from U^{230} are 0.45 and 0.66 Mev, in excellent agreement with our results.

R. Complex Alpha Spectra of Th^{228} and Ra^{224}

Th^{228} (RdTh) is an alpha emitter of 1.9 year half-life.[72] It can be obtained as a daughter product of Ra^{228} by two successive beta emissions. The source of Ra^{228} (MsThI) is natural thorium ores.

Ra^{224} (ThX) is formed by the alpha decay of Th^{228} and has a 3.64 day half-life.[72]

A 67 mc source of Ra^{228} was obtained from the Rare Minerals and Metals Company, Inc., dissolved in 0.6 N HNO_3 and placed on a Dowex 50 ion exchange column. Radium and thorium would both

adhere to the column under these conditions. Then 4 N HNO_3 was passed through the column and most of the radium and actinium would have passed through the column leaving the thorium stuck on the resin. About 1 mc of the thorium was removed from the resin with 1 ml of 16 N HNO_3, evaporated to a few microliters and vaporized from a tungsten filament onto a platinum plate. The sample was exposed for 19 hours in the spectrograph and the alpha spectrum is shown in Fig. 34.

The main group of Ra^{224} was chosen as an energy standard because it had been measured very accurately by Briggs.[2] If the energy of α_o of Ra^{224} is taken as 5.681 Mev, the main group of Th^{228} has an energy of 5.421 Mev. This group is designated α_o of Th^{228} and has an abundance of 71 percent of the total Th^{228} alpha emission. This is in excellent agreement with the value reported by Rosenblum,[73] 5.423 Mev. The second alpha group of Th^{228} is 84.3 kev lower in alpha decay energy than α_o of Th^{228} and is therefore designated α_{84}. Its particle energy was measured as 5.338 Mev in exact agreement with the value reported by Rosenblum.[73] The abundance of α_{84} of Th^{228} was measured as 28 percent of the total Th^{228} alpha emission in exact agreement with the value reported by Rosenblum.[73]

The group at 5.445 Mev is in good agreement with a group at 5.448 Mev, which Rosenblum[74] has attributed to Ra^{224}. We have not determined the intensity of this group accurately as yet, but it appears to be in reasonable agreement with Rosenblum's[74] value of 4.6 percent. Since this group is separated by 240 kev in decay energy from α_o of Ra^{224} it is designated α_{240} of Ra^{224}.

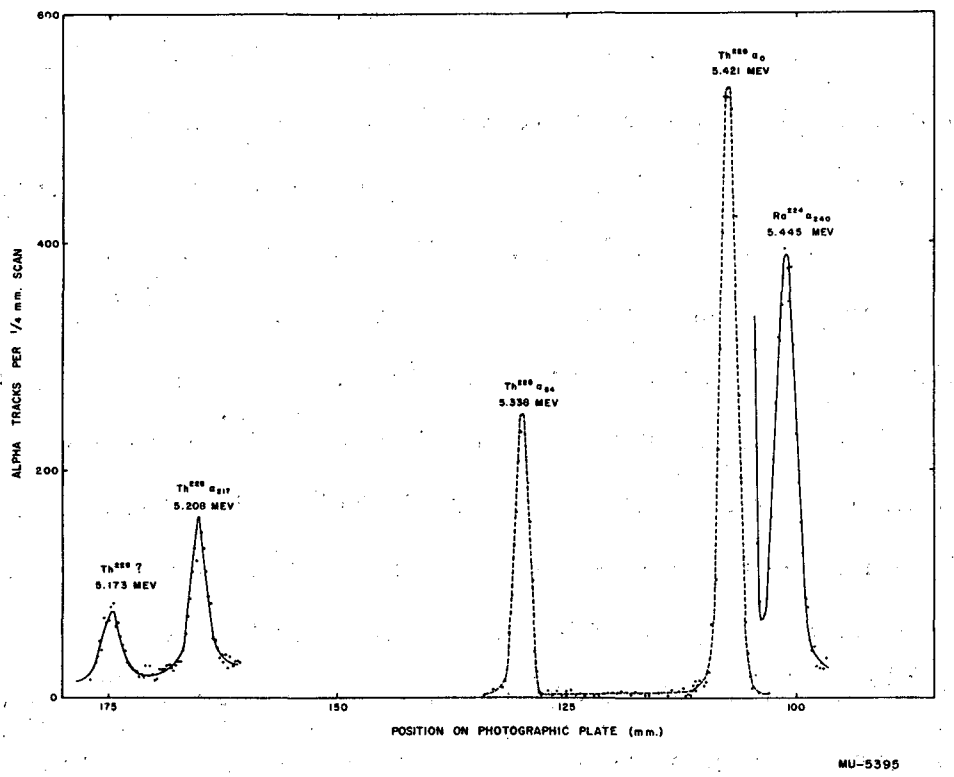

Fig. 34. Th^{228} alpha spectrum.

The groups at 5.208 and 5.173 Mev could have belonged to either Th^{228} or Ra^{224}. In order to decide between these nuclides, another sample of mixed Th^{228} and Ra^{224} activities was prepared. This sample contained 5×10^7 alpha disintegrations per minute and was exposed in the spectrograph for 64 hours. The peak height of α_{240} of Ra^{224} was the same on this exposure as on the run shown in Fig. 34. The peak height of α_{84} of Th^{228} was one-fifth its peak height in the run shown in Fig. 34 and the maximum peak height of any peaks in the region of 5.208 and 5.173 Mev were one-fourth and one-third, respectively, of the values shown in Fig. 34. Therefore, we have concluded that at least the bulk of these alpha groups are emitted by Th^{228}.

The alpha group at 5.208 Mev is separated by 217 kev in decay energy from α_o of Th^{228} and is designated α_{217}. It comprises 0.45 percent of the Th^{228} alpha emission. The 5.173 Mev group is 253 kev in decay energy from α_o of Th^{228} and is designated α_{253}. It comprises 0.22 percent of the Th^{228} alpha emission.

Rosenblum[74] reported an alpha group of Ra^{224} at 5.194 Mev and 0.4 percent abundance. In Fig. 34 there is no alpha group seen at 5.194 Mev greater than 0.1 percent of the Ra^{224} alpha emission.

Another exposure was made with the sample whose spectrum is shown in Fig. 34 at a different magnetic field setting, and no alpha group was observed from 5.145 Mev to 4.797 Mev greater than 0.04 percent of the Th^{228} alpha emission and 0.08 percent of the Ra^{224} alpha emission. We have attributed a group at 4.776 Mev in about 4 percent abundance relative to Th^{228} to Ra^{226} (energy = 4.777 Mev),[75] since MsThI always contains some Ra^{226} and the Th-Ra chemical

purification was not very successful.

A group at 5.297 Mev was also observed in the sample whose spectrum is shown in Fig. 34, but pending further experiments, this group is assumed to be Po^{210} (energy = 5.298 Mev).[4]

S. Gamma Rays of Th^{228}

In order to make a positive identification of the gamma rays of Th^{228}, a quick and quantitative procedure was needed for the separation of Th^{228} from its daughter products. The thorium was placed on a Dowex 50 resin column, heated to 87° C with trichloroethylene, in dilute nitric acid, and the radium removed with 4 N HNO_3. The thorium was stripped off the column rapidly with a one to one volume ratio of lactic acid at a pH of 3. The stripped solution was evaporated down, alpha counted, and pulse analyzed on a 50-channel gamma scintillation counter. The alpha activity of the sample was 3×10^6 alpha dis/min.

The gamma analysis showed four gamma rays due to Th^{228}. The energies determined with Am^{241} 60 kev and U^{235} 184 kev standard were 89, 137, 169 and 212 kev. Since the standards were not run concurrently with the Th^{228}, the energies were susceptible to slight shifts in photomultiplier voltage. In practice it has been found that these shifts amount to less than one channel or 8 kev in this case.

Assuming that one 60 kev Am^{241} gamma ray is associated with 40 percent of the Am^{241} alpha emission, we found the uncorrected respective abundances of the 89, 137, 169 and 212 kev gamma rays to be 1.6, 0.27, 0.12 and 0.22 percent of the Th^{228} alpha emission. After corrections for counting efficiency were applied, the respective

intensities were 1.6, 0.27, 0.14 and 0.32 percent.

The escape peak of the 212 kev radiation was counted as part of the 169 kev radiation, and the escape peak of the 169 kev radiation was counted as part of the 137 kev radiation. The escape peak of the 89 kev radiation comprises 16 percent of the abundance of the peaks. Assuming this ratio applies to all of the peaks, the final intensities for the 89, 137, 169 and 212 kev radiations were 1.6, 0.25, 0.09 and 0.32 percent.

The assignment of these gamma rays to Th^{228} seems certain since the rate at which the 238.6 kev gamma ray of Pb^{212} grew in corresponded to a complete separation of thorium from radium and lead on the ion exchange columns. Since the gamma rays of Bi^{212} were also observed growing in at their prescribed energies, the separation of thorium from bismuth was also effective.

Although it had been supposed for some time that Th^{228} contained two gamma rays between 80 and 90 kev, recent experiments justify the conclusion that only one 83 or 84 kev gamma ray exists. Rosenblum[76] has reported six conversion lines corresponding to an 84.3 kev gamma ray, and Riou[77] has found that only a gamma ray of 83.3 kev belongs to Th^{228}, the other gamma ray of 86.8 kev belonging to one of the daughter products.

The abundance of the 83 kev gamma ray has been determined as 2.1 percent by absorption measurements.[78] Our gamma ray of 89 kev undoubtedly corresponds to the gamma rays mentioned above, the difference in energy being within our experimental error.

1. Decay scheme of Th^{228}. --The gamma ray listed by the workers mentioned above as 83 and 84 kev and measured by us as 89 kev very probably corresponds to the transition from the levels in Ra^{224} populated by α_{84} of Th^{228} to the ground state. The conversion coefficient determined from our work is 17 ± 2. Other workers by absorption methods have found values of 12[79] and ≈ 10.[78] The theoretical L conversion coefficients for E_1, E_2 and M_1 radiation are $0.07 \rightarrow 0.25$, $9 \rightarrow 35$, and $5 \rightarrow 12$, respectively. From our data the multipole order of the transition would be E_2 in agreement with the conclusions by Rosenblum[76] from comparisons of the conversions in the various L shells. The spin of the 84 kev level would therefore be 2 even parity.

The 212 gamma ray must drop from the state in Ra^{224} populated by α_{217} of Th^{228}, since that is the only state populated by sufficient alpha particles to account for the 0.32 percent abundance of the gamma ray. Within experimental error in measuring the gamma energies, the 212 kev radiation agrees with the transition from the 217 level to the ground state. If no other radiation took place from this state and if it were populated by only α_{217} of Th^{228}, the conversion coefficient would be 0.29. The sum of the theoretical K[21] and L[22] conversion coefficients for E_1, E_2 and M_2 radiation would be 0.08, 0.5, and 2.8, respectively. The closest agreement would be with the E_2 radiation. The 137 kev gamma ray, however, probably also descends from the 217 kev level. From its energy and abundance it could descend only from the 253 or 217 kev levels. Since an appreciable portion of the population of the 253 kev level probably

cascades down by the 169 kev radiation, it is deduced that the 137 kev gamma may come from the 217 kev level. The transition would take place between the 217 and 84 kev levels. The total intensity of the 137 and 212 kev radiations is 0.57 percent. Since the alpha population to the 217 kev level is 0.45 percent, the conversion coefficient is very small. The difference between 0.45 percent and 0.57 percent may be due to experimental error. The 212 and 137 kev transitions are therefore E_1 and the 217 kev level has a spin of 1 odd parity.

The energy of the 169 kev gamma ray agrees exactly to the separation between the levels in Ra^{224} populated by α_{253} and α_{84}. The conversion coefficient for this radiation would be 1.4. The sum of $K^{(21)}$ and $L^{(22)}$ theoretical conversion coefficients for E_1, E_2 and M_2 radiation would be 0.1, 1.1, and 4.7. The radiation, therefore, is E_2 and the spin of the 253 kev level is 0, 2 or 4 even parity.

A decay scheme is shown in Fig. 35a.

2. Decay scheme of Ra^{224}. -- Rosenblum[80] has measured K and L conversion lines of a 241 kev gamma ray and assigned it to the transition between the Em^{220} level populated by α_{240} of Ra^{224} and the ground state. A decay scheme is shown in Fig. 35b.

T. Complex Alpha Spectrum of Ra^{223}

Ra^{223} can be obtained from natural uranium by removing and purifying the actinium and then removing the radium daughter that is formed by consecutive beta and alpha decay.

In this study, the radium was removed from the actinium and thorium by first placing the activity on a cold Dowex 50 ion exchange

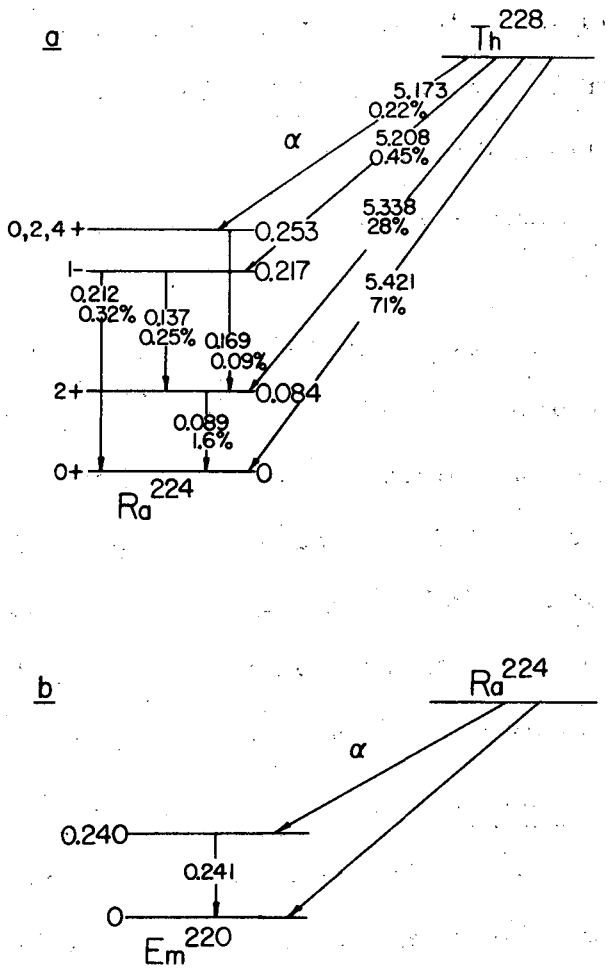

Fig. 35. a) Th^{228} decay scheme.

b) Ra^{224} decay scheme.

column and then eluting with 2 N HNO_3. The Ra^{226} comes off the column before the actinium.

A sample of about 2×10^8 alpha dis/min of Ra^{223} was exposed in the spectrograph. Because the half-life of the emanation gas is too short to be pumped out of the spectrograph before decaying and too long to decay before leaving the area of the spectrograph source, the photographic plates had a tremendous number of random tracks. By a tedious job of alpha track counting a number of alpha groups were resolved. Five groups were observed whose respective particle energy separations from the highest energy group are 0 kev (11 percent), 31 kev (53 percent), 140 kev (25 percent), 207 kev (9 percent), 315 kev (2 percent. These results compare favorably with the work of Rosenblum[81] who found 0 kev (9 percent), 26 kev (53 percent), 134 kev (24 percent), 202 kev (9 percent), 243 kev (2 percent), and 311 kev (3 percent). In addition, Rosenblum reported an alpha group at 5.860 Mev in weak intensity.

No attempt was made to measure the alpha particle energies. In an earlier report the energies were listed using an accepted value of 5.719 Mev[82] for the most intense alpha group. Since that time, the published energy of the most intense alpha group has been changed to 5.704 Mev.[81]

U. Complex Alpha Spectrum of Ra^{226}

Ra^{226} can be obtained in large quantities from natural uranium ores. It is an alpha emitter with a 1622 year half-life.[83]

That Ra^{226} has complex structure could be inferred from the early measurement of a 188-kev gamma ray of Hahn and Meitner.[84]

This gamma ray was shown to be partially converted[85] and the gamma ray energy determined as 186 kev by spectrographic measurement of the conversion electrons.[86]

The alpha particle group for the transition to this excited state was observed with an alpha ray spectrograph by Rosenblum and co-workers,[87] who reported its abundance as ~6.5 percent. The abundance was confirmed as 6.5 percent in another measurement by Bastin-Scoffier.[88] Using an ionization chamber coupled to a pulse-height analyzer, members of this laboratory[89] reported a lower value (4.8 percent) for the abundance of the low energy group. In addition to this well-defined group, Rosenblum and co-workers[87, 88] found some evidence for a weak group of 600 kev lower energy than the main group.

The source employed in the present measurement consisted of 14 micrograms of Ra^{226} as radium chloride sublimed in vacuum onto a 1/8 x 1 in. band on a platinum plate.

The solid curve of Fig. 36 shows the results of a 21-hour exposure for the alpha particle spectrum in the range ~4.5-4.8 Mev with the ordinate showing the number of alpha tracks in each 1/4 mm wide (one field of view of the microscope) scan across the receiver photographic plate. Because of the disparity in track counts for the two peaks, the ordinate scales have been made tenfold different. Because of source thickness, the half-width of the peaks are about 28 kev, as compared with 5-8 kev obtained with other sources in this instrument under best conditions. Nevertheless, the resolution of the two peaks in Fig. 36 is complete, and the abundances of the alpha

groups (5.5 percent $\pm$ 0.2 for the low energy group) should be reliable within the indicated limits of error.

The abundances (Fig. 36) were determined by counting all of the tracks in the low energy peak (almost 26, 000) and those in every fourth 1/4 mm scan for the principal peak (109, 000). The measured abundance of the low energy was 5.6 percent, but this was revised downward to 5.5 percent because of the different geometry factor applicable to the two groups. This difference is a consequence of the longer path followed by the high energy particles because the photographic plate is not normal to the trajectory of the alpha particles but is 60° to the normal. Abundance measurements were also made on three other samples for all of which fewer tracks were registered. The values obtained were 6.0, 5.8 and 5.5 percent for the abundance of the low energy group. The value that we shall adopt is 5.7 $\pm$ 0.3 percent, which is the average of the four measurements and the limit of error encompasses all of them.

In order to illustrate the resolution attainable with thinner sources, an analysis of the main peak was made with a source consisting of only 0.3 microgram Ra^{226}. The peak is shown with a broken line in Fig. 36 and the width at half-maximum is only 6 kev as compared with 26 kev for the thicker source. In this particular measurement the magnetic field was such that the low energy group did not register on the photographic plate.

As mentioned, a weak group of about 600 kev lower energy than the ground state transition has been reported, and its abundance was given in the second publication at 0.17 percent. This group would

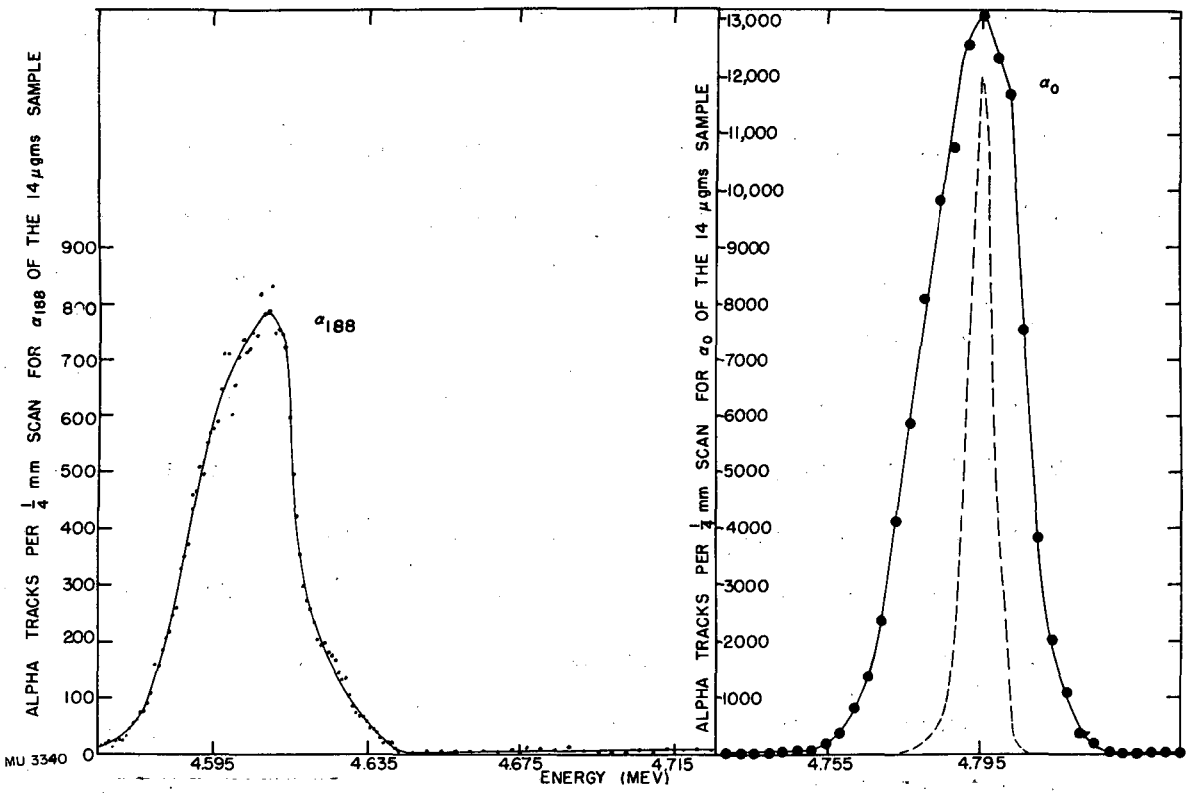

Fig. 36. Ra^{226} alpha spectrum.

placed at 4.20 Mev, and in the present experiments the region from 4.0 to 4.5 Mev was examined. The results are shown in Fig. 37, from which it is deduced that no peak of greater than 0.1 percent abundance could be present.

As seen in Fig. 37, the inability to distinguish groups in lower abundance than 0.1 percent so distant from the main groups is due to an extensive and nearly constant tailing on the low energy side of the peaks. This phenomenon is as yet unexplained. Incidentally, the integrated number of alpha-tracks over this wide range is considerable, but by the same token they must come proportionally from both alpha groups so that the ratio of abundances of the two groups is not affected. Since the limitation of discrimination of an alpha group in this region is not a great deal lower than the reported intensity of the peak α_{600}, a definite disagreement cannot be suggested. However, A. Ghiorso in this laboratory has made a careful examination of Ra^{226} with an alpha particle pulse analyzer over the energy range 3.6-4.4 Mev and set a limit of 0.02 percent for the abundance of any group in this interval.

The energy of α_0 of Ra^{226} was measured as 4.775 Mev, using Po^{210} (5.298 Mev) as a standard. This and the 4.776 Mev value obtained in the experiments on Th^{228} are in excellent agreement with the energy given by Bastin-Scoffier[75] as 4.777 Mev.

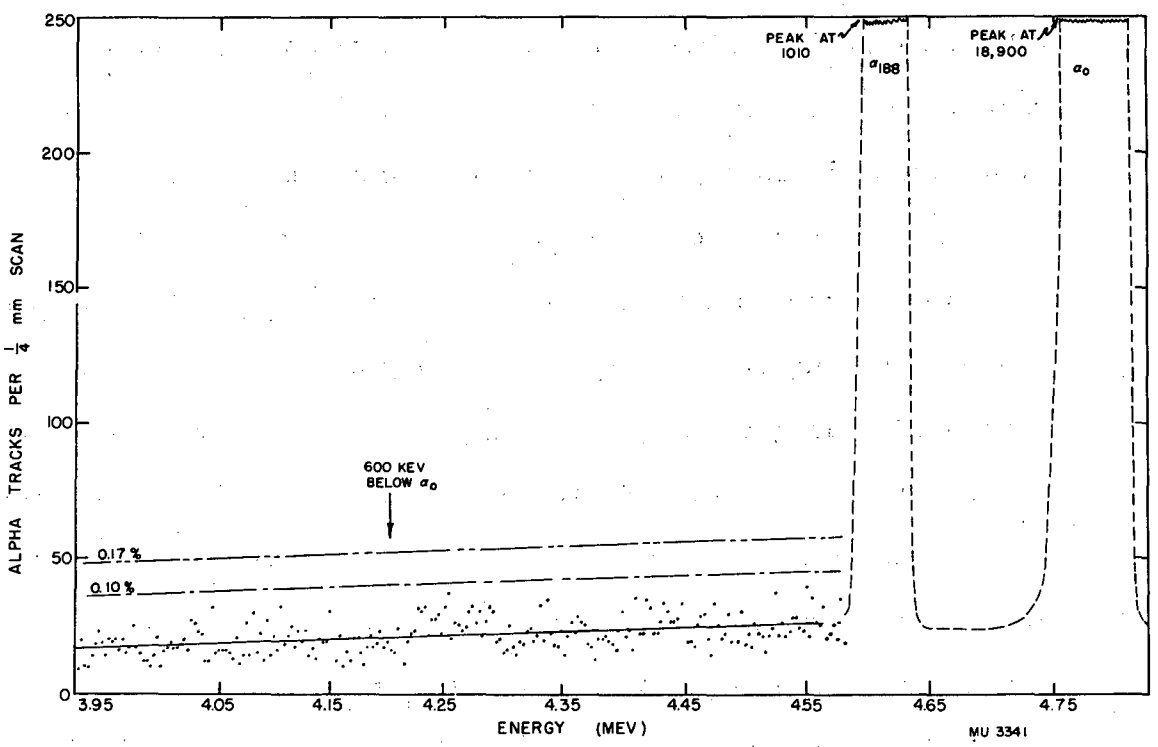

Fig. 37. Ra^{226} low energy alpha spectrum.

..... Alpha tracks per 1/4 mm scan.

— — — Curve indicating the position and the abundances calculated for α_0 and α_{188} from a short exposure.

——— Average background below α_{188}.

— - — A straight line parallel to the average background and 0.10 percent of the α_0 peak height above the average background.

— -- — A straight line parallel to the average background and 0.17 percent of the α_0 peak height above the average background.

V. Complex Alpha Spectrum of Fr^{212}

Francium 212 can be prepared as a thorium spallation product. It has a half-life of 19.3 minutes[90] and 56 percent of the decay goes by electron capture to Em^{212} and 44 percent by alpha emission.[90]

A sample of Fr^{212} adequate for the spectrograph was prepared by vacuum sublimation from a tungsten filament. The sample was exposed in the spectrograph for 3 hours, and a graph of the Fr^{212} and Em^{212} alpha spectra is shown in Fig. 38.

The energy determinations were made using the Em^{212} (6.262 Mev) group as a standard. The group picked as Em^{212} was chosen on the basis of its intensity. Further measurements have been made on the decay rate of the various peaks but the results have not been evaluated.

The most energetic group of Fr^{212} is at 6.408 Mev. Its abundance is about 37 percent of the Fr^{212} alpha emission. This group is designated α_o.

A second alpha group is 22 kev lower in particle energy than α_o and is designated α_{22}. Its alpha particle energy is 6.386 Mev and its abundance is 39 percent.

A third group is found 70 kev lower in particle energy than α_o and is designated α_{71}. Its alpha particle energy is 6.339 Mev and its abundance is 24 percent.

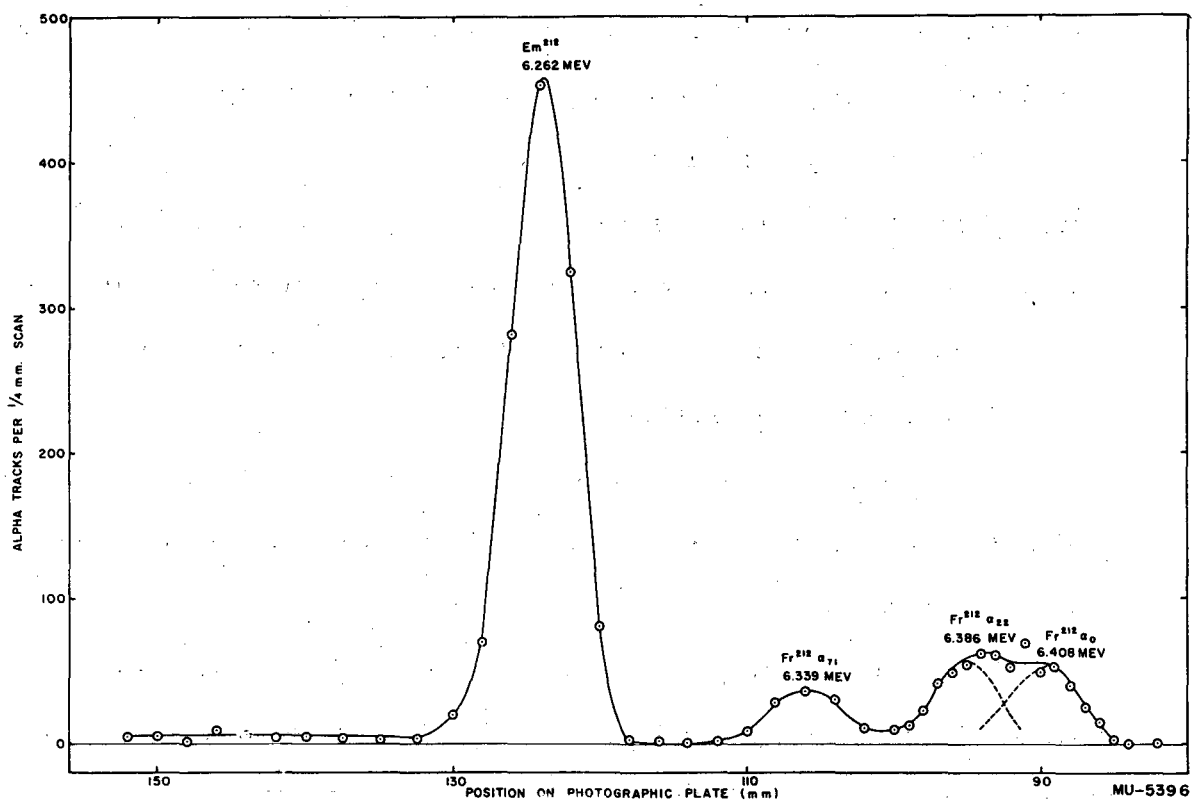

Fig. 38. Fr^{212} alpha spectrum.

W. Complex Alpha Spectra of $Em^{208, 210, 211, 212}$

The light emanation isotopes can be prepared by bombarding thorium with 340 Mev protons.[91] Em^{208}, Em^{210}, Em^{211} and Em^{212} have half-lives of 23 minutes,[92] 2.7 hours,[93] 16 hours,[93] and 23 minutes,[93] respectively.

A sample 5×10^7 alpha c/m was prepared on a 0.0125 in. chromel wire by a glow discharge method.[91]

Five separate exposures of 15 minutes each were made from the sample in order to observe the decay of the various alpha groups. The energies of the groups were determined using as a standard At^{211} (5.862 Mev). The alpha particle energy of Em^{208} was 6.138 Mev. This compares with the ion chamber value of 6.14 ± 0.02 Mev.[91]

The alpha particle energy of Em^{210} was 6.036 Mev. This compares with the ion chamber value of 6.02 ± 0.02 Mev.[93]

The most energetic alpha group of Em^{211} had a particle energy of 5.847 Mev and an abundance of 33 percent. A second alpha group of Em^{211} had an energy of 5.778 Mev and is designated α_{70}. Its abundance is 67 percent.

The alpha particle energy of Em^{212} was 6.262 Mev. This compares with ion chamber measurements of 6.23 ± 0.02 Mev.[93]

All of the above measurements were made by determining peak separations from extrapolation of the high energy edges down to the background.

X. Alpha-Gamma Coincidences of Em^{211}

Alpha-gamma coincidence measurements were made on Em^{211} with the single-channel analyzer described in an earlier section. A gamma ray of 72 kev was found to be in coincidence with alpha particles in the Em^{211} sample. By observing the decay of the coincidence peak it was determined that the At^{211} and Po^{211} could not be causing the coincidences. Since the coincidences decayed with a 16-hour half-life, the 72 kev gamma was assigned to Em^{211}.

The conversion coefficient of the 72 kev gamma assuming it belongs to Em^{211} was about 60. This radiation probably represents the transition between the levels in Po^{207} populated by α_{70} and α_0 of Em^{211}.

Y. Complex Alpha Spectra of At^{211}, Po^{211} and At^{210}

At^{210} and At^{211} can be prepared by bombarding bismuth with alpha particles. At^{211} has a half-life of 7.5 hours[94] and decays 59.1 percent by electron capture and 40.9 percent by alpha emission.[95] Po^{211} is formed by the electron capture decay of At^{211} and decays by alpha emission with a 0.52 second half-life.[96] At^{210} has a half-life of 8.3 hours[97] and decays mainly by electron capture with some slight alpha branching.[98]

A number of samples of At^{211} and mixed astatine isotopes were prepared by subliming the astatine directly from the bismuth target onto a platinum plate. Only one alpha group of At^{211} was observed. Its energy, determined against Am^{241} and Po^{218} standards, was 5.862 Mev. From ion chamber measurements values of 5.85[91] and 5.89 Mev[99] have been reported.

A graph of the spectrum of a sample containing mixed astatine isotopes is shown in Fig. 39. Three positive alpha groups of At^{210} were found at 5.519 (32 percent), 5.437 (31 percent), and 5.355 (37 percent) Mev. These energies were determined using Po^{210} (5.298 Mev) as an internal standard. Using the Po^{210} as a criterion of the amount of At^{210} electron capture, we found 0.17 percent of the At^{210} decay is by alpha emission. The Po^{210} peak was identified as such since it did not decay noticeably in one day.

Besides the main group, the Po^{211} spectrum showed two alpha groups at 6.88 (0.50 percent) and 6.56 (0.53 percent) Mev, in excellent agreement with the previous results of 6.90 (0.6 percent) and 6.57 (0.5 percent) Mev.[96] A group reported at 6.34 Mev in 0.1 percent abundance was not observed. Its maximum intensity would have been 0.02 percent. The limits of error for the energy of the 6.34 Mev group, however, were given as 60 kev, and our measurements did not encompass the entire range of the limits of error.

Z. Alpha Spectra of Po^{208} and Po^{209}

The alpha particle energies of Po^{208} and Po^{209} were determined as 5.108 and 4.874 Mev, respectively, using Po^{210} as a standard. A graph of these spectra is shown in Fig. 40 to indicate the absence of any complex structure in Po^{208}.

From electron capture of Bi^{204}, the first excited state of Pb^{204} is deduced as 384 kev.[100] The maximum intensity of any alpha group of Po^{208} corresponding to this energy difference is 0.016 percent. Other values for the particle energy of Po^{208} are 5.109 Mev,[101] and 5.10 Mev,[102] while another value for Po^{209} is 4.86 Mev.[102]

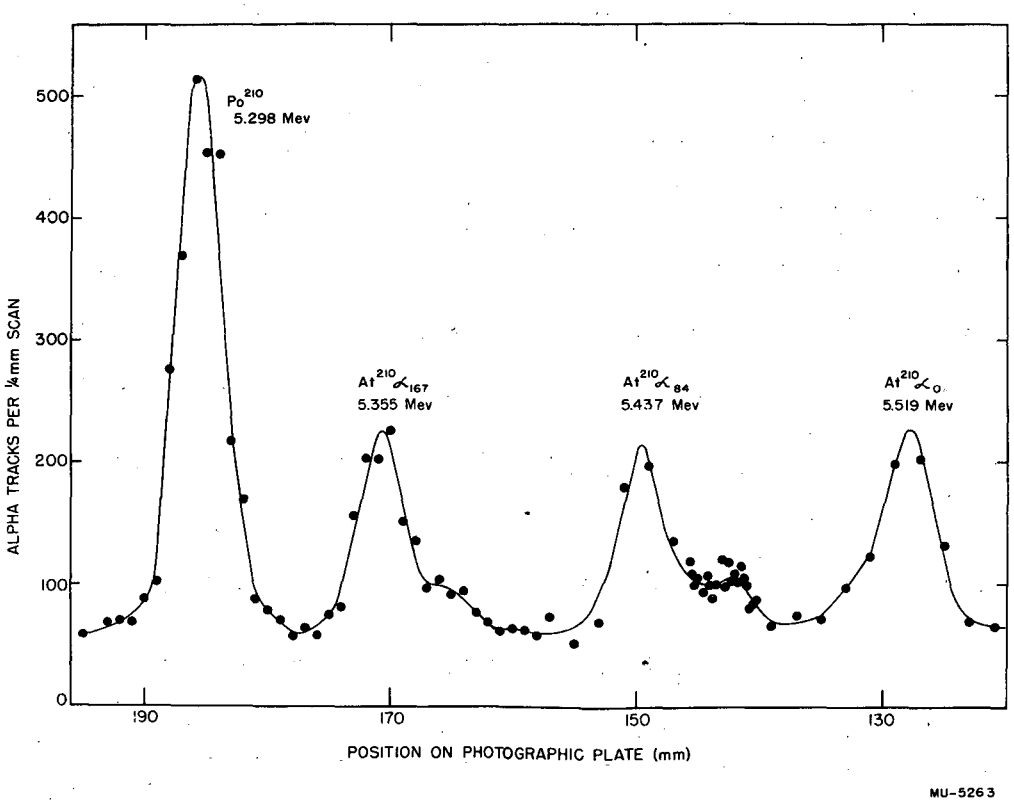

Fig. 39. At^{210}, Po^{210} alpha spectra.

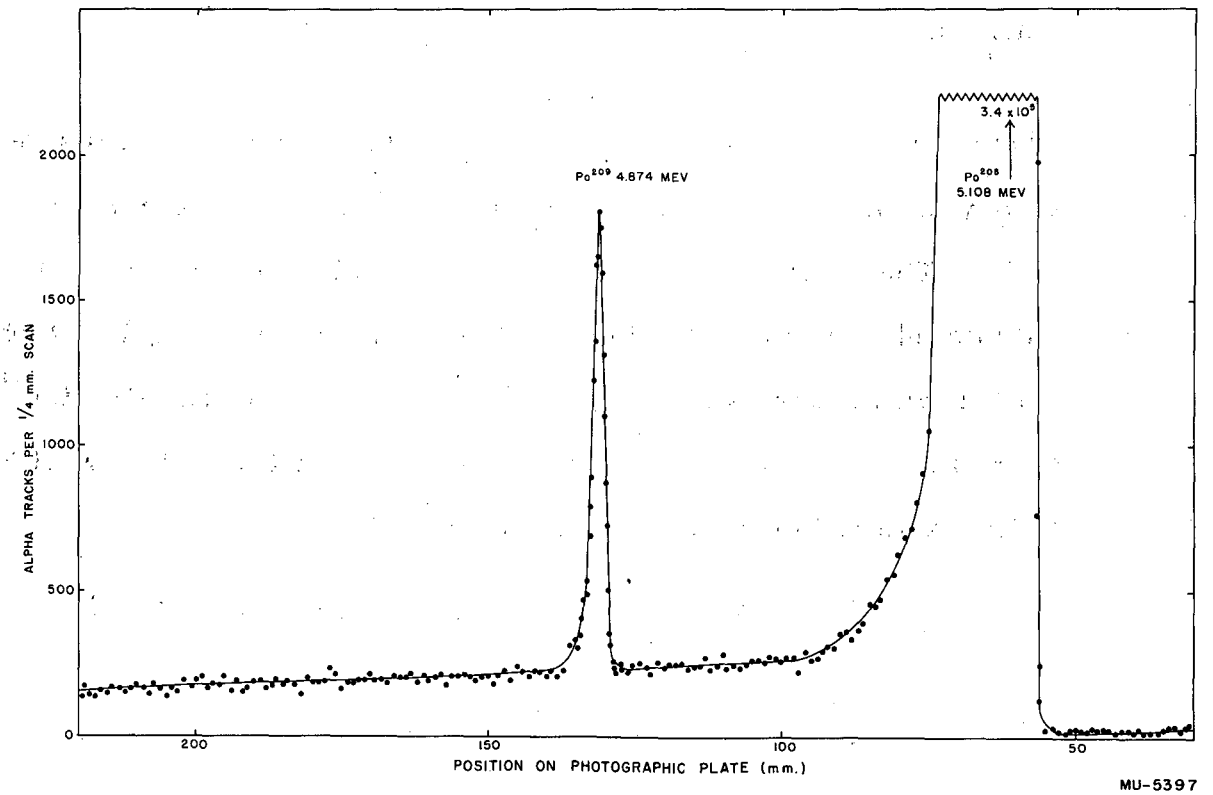

Fig. 40. $Po^{208, 209}$ alpha spectra.

III. DISCUSSION OF RESULTS

A. Ground State Transitions of the Even-Even Nuclei

A number of equations have been presented in the literature relating the half-life, charge, energy, mass and radius of even-even nuclei. The only parameter which cannot be measured explicitly is the nuclear radius. The validity of the various equations is determined by the invariance of the calculated parameter, r_o, in the equation

$$r_o = r/A^{1/3},$$

where r is the nuclear radius calculated from one of the alpha decay theory equations and A is the mass of the daughter nucleus.

The radii of the various even-even alpha emitters were calculated from the best data available and are shown in Table 16. The equation used was the Kaplan[103] adaptation of the Preston[104] formula. The data not obtained in this research were taken from "The Table of Isotopes" (see reference 27).

$$\lambda = \frac{2v}{r} \frac{\mu^2 \tan \alpha}{\mu^2 + \tan^2 \alpha} e^{-\frac{8\varepsilon^2 Z}{\hbar v}(\alpha - \sin \alpha \cos \alpha)}$$

$$\mu = \left(\frac{0.52}{E}\right)^{1/2} \qquad \cos \alpha = \left(\frac{mv^2 r}{4\varepsilon^2 Z}\right)^{1/2}$$

Z = charge of the daughter nucleus

m = reduced mass

v = alpha velocity $= \left(\frac{2E}{M}\right)^{1/2}$

e = the electrostatic unit of charge

$\hbar = \frac{h}{2\pi}$ h = Planck's constant

The energy, E, is the sum of the alpha particle energy, the nuclear recoil energy, and a correction of the type described by Ambrosino and Piatier[105] which takes into account the orbital electrons in the nucleus. The values of this correction which we used are shown in column 3 of Table 16 and were deduced by Dr. J. O. Rasmussen of this laboratory from an article by Dickinson.[106]

In Table 17 are shown the values of r_o for the nuclides with the best known energy and half-life. It is seen that as Z increases from emanation to curium, the value of r_o decreases by about 4 percent. This is outside the experimental error of the measurements.

A reconsideration of the data in Table 16 shows that for a given element the value of r_o is constant to 1 percent except for Pu^{242}, U^{238}, Th^{232}, and the U^{230} series. Measurements with the alpha particle spectrograph indicate that the energy of U^{230} α_o may be 5.89 instead of 5.85 Mev. This would bring the respective values of r_o for U^{230}, Th^{226}, and Ra^{222} into close agreement with the other isotopes. The energies of U^{238} and Th^{232} are not known with any accuracy since ion chamber measurements in this region are unreliable. One might predict that the listed energy values are low by about 30 kev. The half-life of Pu^{242} is not known with accuracy and this might account for the rather large deviation of its r_o from the other plutonium isotopes.

Table 16
Radii of Even-Even Nuclei

Parent nucleus	Alpha particle energy (Mev) used in calculations	Ambrosino-Piatier correction	Experimental half-life	Experimental partial half-life of highest energy alpha group	Radius 10^{-13} cm	Radius $A^{1/3} 10^{-13}$ cm r_o	Calculated partial half-life
Cm^{244}	5.798	0.040	19 y	25.3 y	9.32	1.500	25.3 y
Cm^{242}	6.110	0.040	162.5 d	220.5 d	9.32	1.504	222 d
Pu^{242}	4.898	0.039	9×10^5 y	1×10^6 y	9.22	1.488	1.05×10^6 y
Pu^{240}	5.162	0.039	6580 y	8.66×10^3 y	9.41	1.522	8.67×10^3 y
Pu^{238}	5.492	0.039	89.6 y	118 y	9.33	1.514	119 y
Pu^{236}	5.75	0.039	2.7 y	3.37 y	9.39	1.528	3.31 y
U^{238}	4.18	0.037	4.49×10^9 y	5.76×10^9 y	9.55	1.550	5.75×10^9 y
U^{236}	4.499	0.037	2.39×10^7 y	3.27×10^7 y	9.36	1.524	3.20×10^7 y
U^{234}	4.767	0.037	2.48×10^5 y	3.35×10^5 y	9.37	1.529	3.34×10^5 y
U^{232}	5.31	0.037	70 y	100 y	9.35	1.531	101 y
U^{230}	5.85	0.037	20.8 d	27.0 d	9.48	1.557	26.6 d
Th^{232}	3.98	0.036	1.39×10^{10} y	1.83×10^{10} y	9.54	1.562	1.84×10^{10} y
Th^{230}	4.682	0.036	8.0×10^4 y	1.07×10^5 y	9.35	1.535	1.06×10^5 y
Th^{228}	5.423	0.036	1.90 y	2.64 y	9.32	1.535	2.63 y
Th^{226}	6.30	0.036	30.9 m	39.6 m	9.42	1.556	40.0 m
Ra^{226}	4.777	0.035	1622 y	1.72×10^3 y	9.35	1.545	1.74×10^3 y
Ra^{224}	5.681	0.035	3.64 d	3.83 d	9.33	1.546	3.89 d
Ra^{222}	6.51	0.035	38 s	---	9.42	1.566	38 s
Em^{222}	5.486	0.034	3.825 d	---	9.34	1.552	3.80 d
Em^{220}	6.282	0.034	54.5 s	---	9.36	1.561	54.2 s
Em^{218}	7.12	0.034	0.019 s	---	9.52	1.591	0.019 s

Table 16 (Cont'd)
Radii of Even-Even Nuclei

Parent nucleus	Alpha particle energy (Mev) used in calculations	Ambrosino-Piatier correction	Experimental half-life	Experimental partial half-life of highest energy alpha group	Radius 10^{-13} cm	Radius $A^{1/3}10^{-13}$ cm r_o	Calculated partial half-life
Po^{218}	5.998	0.033	3.05 m	---	9.23	1.543	3.04 m
Po^{216}	6.774	0.033	0.158 s	---	9.20	1.543	0.152 s
Po^{214}	7.680	0.033	1.637×10^{-4} s	---	9.18	1.545	1.64×10^{-4} s
Po^{212}	8.776	0.033	3.04×10^{-7} s	---	9.05	1.527	3.02×10^{-7} s
Po^{210}	5.298	0.033	138.3 d	---	8.40	1.423	138.1 d
Po^{208}	5.108	0.033	2.93 y	---	8.49	1.442	2.93 y
Po^{206}	5.21	0.033	9 d	90 d	8.77	1.495	90.0 d

Table 17

Radii of Even-even Nuclei for Which Precision Energies and Half-lives Are Available

Parent Nucleus	r_o
Cm^{242}	1.504
Pu^{240}	1.522
Pu^{238}	1.514
Th^{230}	1.535
Th^{228}	1.535
Ra^{226}	1.545
Ra^{224}	1.546
Em^{222}	1.552
Em^{220}	1.561
Po^{218}	1.543
Po^{216}	1.543
Po^{214}	1.545

B. Alpha Transitions to the First Excited State of Even-Even Nuclei

In Table 18 are seen the energies of the first excited state as detected by alpha decay measurements. Included in the table are results from beta decay and electron capture in the region of interest. In Fig. 41 these data are plotted on a graph of energy versus neutron number. The data in Table 18 for which no references are given were taken from this research or from "The Table of Isotopes" (see reference 27).

The partial half-lives of the alpha transitions to the first excited state were calculated by the Kaplan formula mentioned earlier using the nuclear radius determined for the ground state transition. Table 19 shows a list of the ratios of experimental half-life divided by the theoretical half-life. These ratios are shown in Fig. 42. A similar plot has been made by another worker.[111] These ratios are about equal to one or larger. The average of the ratios for a given element seems to reach a minimum in the region of thorium and then rises at higher and lower atomic numbers. It is striking that the minimum ratio for the alpha decay of Po^{208} to Pb^{204} is 32. This is especially noticeable since the ratios for Po^{210} and Po^{206} are quite low.

In two cases, Cm^{242} and Pu^{238} alpha decay, the spin of the first excited state was measured and found equal to 2 even parity in agreement with expectations.

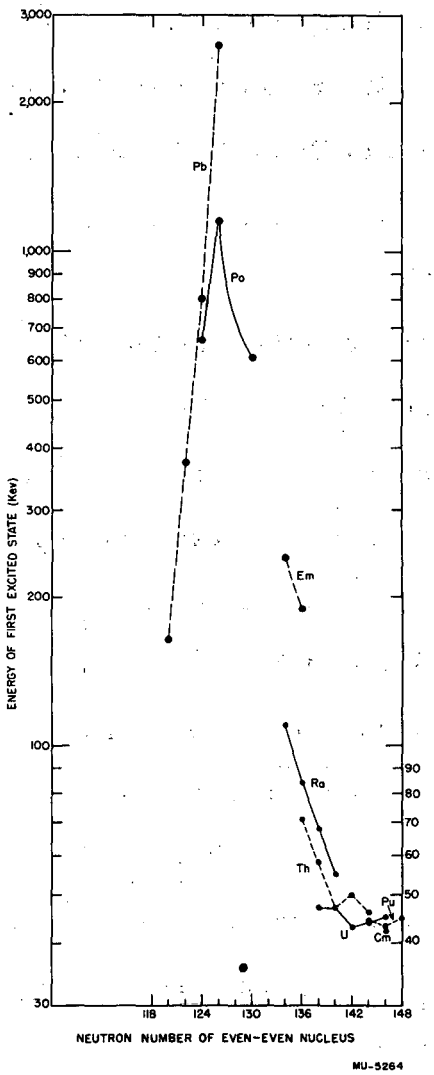

Fig. 41. Energies of first excited states of even-even nuclei.

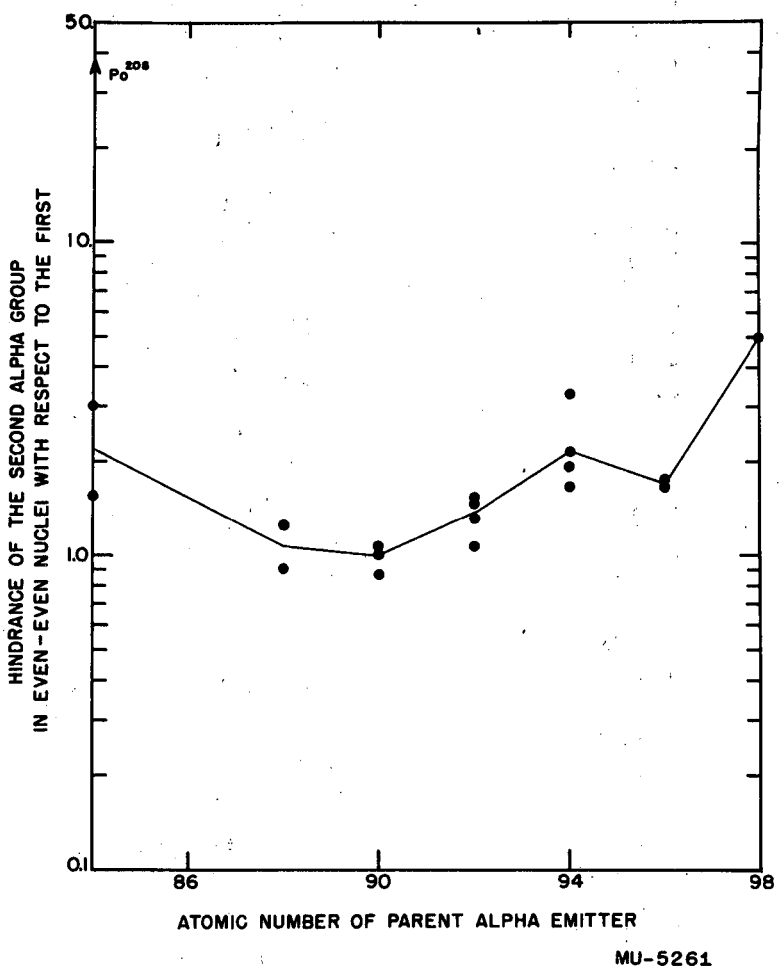

Fig. 42. Relation of relative intensity of alpha group to the first excited state of even-even nuclei with respect to alpha decay theory.

Table 18

Energies of First Excited States of Even-Even Nuclei

Nuclide	Energy of First Excited State (Mev)	Parent of Nuclide and Type of Decay	Reference
Cm^{242}	0.042	Am^{242} (16 h) β^-	(107)
		Cf^{246} α	(108)
Pu^{242}	0.045	Am^{242} (16 h) e.c.	(107)
Pu^{240}	0.043	Cm^{244} α	
Pu^{238}	0.044	Cm^{242} α	
U^{238}	0.045	Pu^{242} α	
U^{236}	0.044	Pu^{240} α	
U^{234}	0.043	Pu^{238} α	
U^{232}	0.045	Pu^{236} α	
U^{230}	0.047	Pu^{234} α	(108)
Th^{234}	0.045	U^{238} α	
Th^{232}	0.050	U^{236} α	
Th^{230}	0.050	U^{234} α	
Th^{226}	0.071	U^{230} α	
Ra^{228}	0.055	Th^{232} α	
Ra^{226}	0.068	Th^{230} α	
Ra^{224}	0.084	Th^{228} α	
Ra^{222}	0.110	Th^{226} α	
Em^{222}	0.188	Ra^{226} α	
Em^{220}	0.240	Ra^{224} α	
Po^{214}	$\gtrsim$ 0.609		
Po^{212}	$\gtrsim$ 0.72	Bi^{214} β^-	
Po^{210}	1.15	At^{210} (e.c.)	
Po^{208}	0.66	At^{208} (e.c.)	(109)
Pb^{208}	2.6143	Tl^{208} β^-	
Pb^{206}	0.800	Po^{210} α	
Pb^{204}	0.374	Bi^{204} e.c.	
Pb^{202}	0.163	Po^{206} α	(110)

Table 19

Ratio of Experimental to Theoretical Half-life for Alpha Emission to the First Excited State of Even-even Nuclei

Parent alpha emitter	$\frac{T_{1/2}\ \text{Experimental}}{T_{1/2}\ \text{Theoretical}}$	Energy of first excited state used in calculation
Cf^{246}	5	0.04
Cm^{244}	1.8	0.043
Cm^{242}	1.7	0.044
Pu^{242}	2.2	0.045
Pu^{240}	1.7	0.044
Pu^{238}	1.9	0.043
Pu^{236}	2.2	0.047
Pu^{234}	3.5	0.047
U^{238}	1.5	0.045
U^{236}	1.1	0.050
U^{234}	1.3	0.047
U^{232}	1.1	0.058
U^{230}	1.5	0.069
Th^{232}	1.0	0.055
Th^{230}	1.0	0.068
Th^{228}	0.9	0.084
Th^{226}	1.1	0.117
Ra^{226}	0.9	0.188
Ra^{224}	1.25	0.237
Po^{210}	1.6	0.800
Po^{208}	>32	0.374
Po^{206}	3.1	0.163

C. Alpha Transitions to Higher Excited States of Even-Even Nuclei

In Fig. 43 the ratio of the energy of the various excited states to the first excited state as a function of neutron number of the daughter nucleus is plotted. From gamma ray data on the alpha decay of Cm^{242} and Pu^{238} it was observed that the second excited state decayed to the first excited state. For U^{234} and Th^{230} alpha decay, the energy of the second excited state was taken as equal to the sum of the gamma ray corresponding to decay of the first excited state and the next highest energy gamma. In the case of Th^{230}, gamma rays of 68, 140 and 240 kev were observed by Curie;[112] 70 ± 2, 145 ± 3, and 235 ± 5 kev by Rasetti and Booth;[113] and 69, 146 and 246 kev by ourselves. Rosenblum[114] reported conversion lines of a 67.8 kev gamma ray and an electron line corresponding to K conversion of a 228 kev gamma ray. This latter line could correspond, however, to an L_2 conversion of a 141 kev gamma ray.

A recent theoretical argument in the literature by Aage Bohr[115] predicted that the energies of the excited states of a given nucleus should be proportional to $J(J + 1)$, where J = the total angular momentum of the states. The values of J were restricted to even spins and even parity. The expected ratio of the energy of the second excited state to the first excited state would then be $\left[4(4 + 1)\right] / \left[2(2 + 1)\right] = 3.3$. It is seen from Fig. 43 that this is in excellent agreement with the experimental results above Th^{228} alpha decay. The spins of the second excited states of Pu^{238} and U^{234} are probably 0, 2 or 4 even parity. The spin of the second excited state

of Pb^{208} appears to be 4 even parity from analysis of its K conversion coefficient and the lack of any cross-over transition. If the decay scheme for U^{234} alpha decay is correct, the second excited state of Th^{230} has a spin of 0, 2 or 4 even parity.

In the case of Th^{228} alpha decay, a state of spin 0, 2 or 4 even parity is found whose energy ratio to the first excited state is 3.0, in good agreement with the examples at higher neutron numbers. An additional state is found, however, with a spin of one odd parity. Recent tentative experiments have shown that U^{230} and Th^{226} have two observable low intensity groups like Th^{228}. Since the "extra" alpha group appears in Th^{228}, Th^{226} and U^{230} alpha decay, but not in Th^{230} or U^{232} alpha decay, it appears to be a function of the number of neutrons in the nucleus. The daughters of Th^{228} and U^{230} have 136 neutrons. If the next shell that fills after the closed shell of 126 neutrons is the g 9/2 instead of the i 11/2, then 136 neutrons represent a closed sub-shell. This may be a reason for the distinct change in nature of the alpha emission from 136 to 138 neutrons.

Aage Bohr[107] also stated that as one approaches a closed shell, i.e., 126 neutrons, the $J(J+1)$ relation might break down.

Some unclassified gamma rays in Cm^{242}, Pu^{238} and Th^{230} remain. If the energy of these gamma rays is divided by the energy of the respective first excited states, the result is very nearly a constant. In addition, if one adds these energies to the respective energies of the second excited states, the ratio of the resulting energies to the energy of the first excited state is nearly equal to 7 as is shown in Fig. 43. Recalling the $J(J+1)$ relation, we find the

expected ratio of the energy of the third excited state and first excited state is $[6(6+1)] / [2(2+1)] = 7$.

The half-lives of the alpha emission to the second excited state were calculated from the Kaplan formula using the radius calculated for the ground state transition. The log of the ratio of the experimental half-life to the calculated half-life is plotted in Fig. 44, versus atomic number.

The third and fourth alpha groups of Th^{228} have about the same ratio of experimental half-life to calculated half-life. If the spins of the second excited states of the nuclei above Ra^{226} are 4 as predicted by Bohr, then there must be sizeable factors capable of hindering the alpha decay process besides spin.

D. Alpha Transitions of Odd Mass Nuclei

These nuclei do not exhibit the same degree of regularity as even-even nuclei. The partial half-lives of the ground state transition cannot be calculated with any accuracy, and the ratios of the intensities of various alpha groups appear to vary almost indiscriminately.

Some regularities have been observed. Lower in energy than the most intense alpha group by about the separation expected for the first excited state of even-even nuclei in that region is another alpha group. The intensity of this alpha group relative to the main group is roughly the same as for even-even nuclei in this region. This effect has been observed for Cm^{243}, Am^{243}, Am^{241}, Pu^{241}, Pu^{239} and U^{233}. In Am^{241}, Pu^{239} and Am^{241}, the state populated by the lower energy group of this "even-even" pair, decays by a

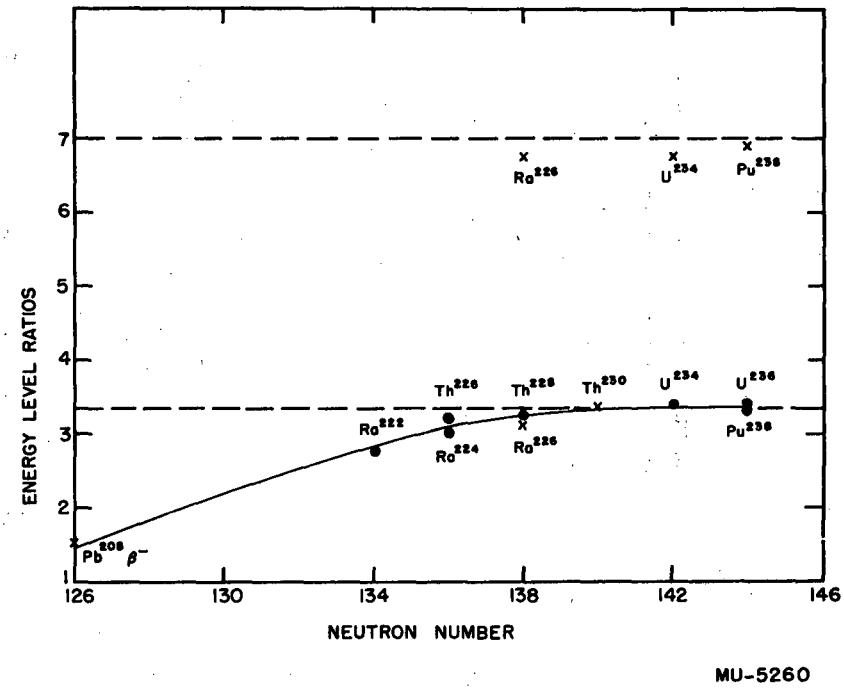

Fig. 43. Ratios of energies of second and higher excited states to the first excited state of even-even nuclei.

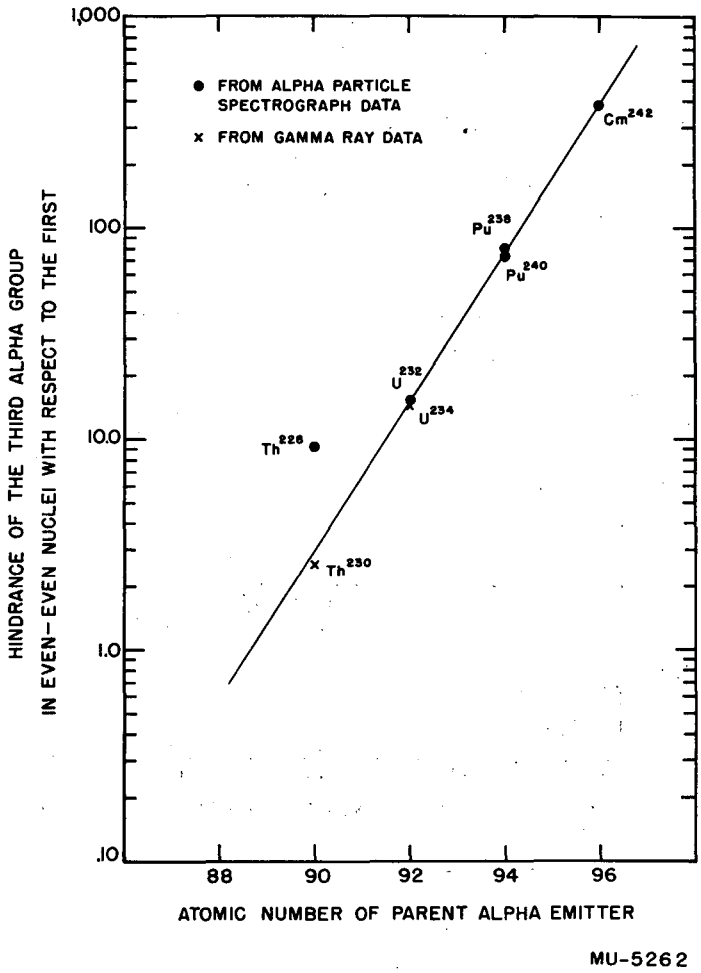

Fig. 44. Relation of relative intensity of alpha group to the second excited state of even-even nuclei with respect to alpha decay theory.

highly converted transition to the state populated by the higher energy alpha group. The position of this pair may vary from very close to several hundred kev above the ground state. A possible explanation for these "even-even" characteristics may lie in the concept of decay of the even-even core of the parent nucleus with a minimum rearrangement of the odd nucleon in the nucleus.

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V. APPENDIX I

An even-even nucleus is generally considered to have 0 spin and even parity. If this nucleus decays by alpha emission, the product of the intrinsic parities of the daughter products and the parity associated with the nuclear reaction must be even in order to conserve parity. Since the parity of one of the daughter products, the alpha particle, is even, then the product of the parity of the daughter nucleus and the parity associated with the nuclear reaction must be even.

If the alpha particle leaves the nucleus with an odd number of units of angular momentum, the spin of the daughter nucleus will be odd since the alpha particle has 0 intrinsic spin. The parity

associated with a nuclear reaction involving a change of an odd number of units of angular momentum is odd. Therefore the parity of the daughter nucleus must be odd.

If the alpha particle leaves the nucleus with an even number of units of angular momentum, the spin of the daughter nucleus will be even. The parity associated with a nuclear reaction involving a change of an even number of units of angular momentum is even, therefore the parity of the daughter nucleus is even.

Therefore, any state of an even-even nucleus populated by alpha decay will have even spin and even parity or odd spin and odd parity.

From gamma ray selection rules, transitions involving a change of an even number of units of angular momentum and no parity change have an electric multipole order. Likewise, transitions involving a change of an odd number of units of angular momentum and parity change have an electric multipole order. Therefore, all gamma ray transitions from an even-even nucleus excited state populated by alpha decay to the ground state are electric.

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