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ABSTRACT

The gamma rays of AcK (Fr^{223}) were studied in a sodium iodide-photomultiplier scintillation counter coupled to a 50 channel pulse height analyzer. Photons of a 49.8 kev gamma ray appear in 40 ± 10 percent and photons of an 80 kev gamma ray appear in 24 ± 6 percent of the total beta disintegrations. In addition, low intensity gamma rays appear at 215 and 310 kev. L x-radiation is present in about the same intensity as the 49.8 kev gamma ray. Gamma-gamma coincidence studies proved that the 49.8 and 80 kev radiations were not in coincidence. It was also shown that the 215 and the 50 kev gamma rays are not in coincidence. The L x-radiation was studied in a proportional counter coupled to a 50 channel analyzer and identified as radium x-rays. Since Th^{227} decays to the same daughter as does AcK some measurements were made on its gamma spectrum. Prominent photopeaks were observed at 49.8, 87, and 235 kev. The 235 and 49.8 kev gamma rays were observed to be in coincidence. The known decay data for AcK and Th^{227} cannot be fitted into a common decay scheme without additional information.

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I. INTRODUCTION

AcK (Fr^{223}) is a member of the U^{235} decay chain in which it appears as a result of a 1.2 percent alpha branching in the decay of Ac^{227}. AcK decays primarily by beta emission with a half-life of 21 minutes into 11.2-day AcX (Ra^{223}), although recently a very slight alpha branching ($\sim 4 \times 10^{-5}$) has been reported.[1] Investigations of the gamma radiations of AcK have been carried out by Lecoin and co-workers[2] using absorption techniques. The present report concerns a more detailed study of these radiations using a scintillation crystal spectrometer and a proportional counter in combination with a multichannel pulse height analyzer and also using gamma-gamma coincidence techniques.

II. PREPARATION AND STANDARDIZATION OF SAMPLES

A stock solution of Ac^{227} dissolved in 3 ml of saturated hydrochloric acid was used as a source of francium. Whenever a fresh sample of AcK was required one drop of a 0.4 M silicotungstic acid solution was added to the ice cold Ac^{227} solution. Silicotungstic acid precipitated, carrying the francium, and was centrifuged out. The precipitate was dissolved in

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distilled water and reprecipitated by saturating the solution with hydrogen chloride gas. This process was repeated two or three times. A water solution of the last silicotungstic acid precipitate was quickly passed through a 1 cm x 4 mm column of Dowex-50 ion exchange resin. The adsorbed AcK was washed free of silicotungstic acid by 2 ml of distilled water, then stripped from the column with 300 µl of 10 M hydrochloric acid. Aliquots of this solution were evaporated on platinum counting discs. These samples were weightless and of high radiochemical purity. Further details on this preparation method are given elsewhere.[3]

The samples were all counted with a geiger counter using the third shelf of the geiger tube mount standard to this laboratory. The tube used was an Amperex geiger tube filled with a mixture of argon and chlorine. An aliquot of each preparation was followed for decay as a check on radiochemical purity. The observed counting rate was translated into disintegrations per minute by multiplying by the calibration factor, 42.

This calibration factor was based on the determination of the alpha counting rate of the AcK daughters and was determined as follows. Samples of AcK isolated as described above were mounted on platinum plates. Asbestos washers were placed above the samples and a clean collector platinum plate was placed above. By touching a torch to the bottom plate for one second and bringing the platinum to a red glow for this instant the francium was volatilized to the collector foil leaving behind any traces of Ac^{227}, AcX or RdTh present at that time. This instant was taken as zero time. The geiger counting rate of the volatilized francium sample was followed and back extrapolated to zero time. Several hours later the alpha disintegration rate of the sample was carefully measured in an alpha

counter (52 percent geometry). One-fourth of this alpha activity was ascribed to the 11.2 day AcX daughter of AcK and by straightforward calculations the initial disintegration rate of AcK was determined. This method of standardization substitutes the uncertainties of alpha counting, which are considerably less, for the uncertainties of absolute beta counting arising out of the back scattering, window absorption and other corrections. The purity of the alpha activity was checked by analysis of the alpha spectrum. This was done by introducing to a 48 channel differential pulse height analyzer the alpha pulses developed in a large ionization chamber after suitable linear amplification. This analysis was performed by A. Ghiorso on equipment developed by him as an improvement of apparatus previously described.[4] The alpha spectrum curves verified that the alpha activity was pure Ra^{223} plus daughters with no contribution from Th^{227}.

III. SPECTROMETER EQUIPMENT

The spectrometer equipment used in this work was assembled by A. Ghiorso and A. E. Larsh of this laboratory. The gamma detection initially occurred in a 1.5-inch diameter by 1-inch thick crystal of sodium iodide (thallium-activated) procured from the Harshaw Chemical Company mounted below a Dumont 6292 tube using a method similar to that described by Borkowski.[5]

On the side of the crystal facing the photomultiplier tube was affixed a quartz disc; a layer of oil between the quartz and the outside surface of the tube provided optical coupling. The other surfaces of the tube were packed with a reflecting layer of magnesium oxide. The whole

assembly was mounted in an aluminum lined lead shield on top of a standard geiger tube 5 position shelf assembly. Incident gamma rays penetrated a thin foil of beryllium (~150 mg/cm^2) and a thin layer of magnesium oxide (about 1/16 inch) before entering the crystal.

The output pulse from the photomultiplier was amplified in a pre-amplifier, then in a linear amplifier. The final pulse was introduced to a 50 channel differential pulse height analyzer. This analyzer, based on a novel use of a 6BN6 as one arm of a gated univibrator, is a new design of Ghiorso and Larsh.[6] After proper alignment the channel width stability (operating at a 5 volt channel width) was better than 1 percent and remained so for periods of weeks. Gain and bias controls permitted the inspection of any predetermined energy interval with the full 50 channels. Further details on this equipment will be obtainable in a forthcoming publication.[6]

For measurements on the beta spectrum the sodium iodide detector was replaced by an anthracene crystal 1.5 inches in diameter by 0.25 inch thick mounted in front of an RCA-5819 photomultiplier tube.

A proportional counter was used to study L x-rays. An Eck and Krebs tube 19 cm in diameter was filled to one atmosphere with a 90 percent xenon-10 percent methane mixture and operated at 1100 volts. After preamplification the pulse from the proportional counter was introduced to the linear amplifier of the 50 channel analyzer.

For study of gamma-gamma coincidences the apparatus assembled by A. E. Larsh and F. Asaro was used. In this equipment the sample was mounted between two sodium iodide crystals 1.5 inches in diameter and 1 inch thick with the front edge of the two crystals only 0.25 inch apart

for maximum geometry. Each crystal had its own Dumont 6292 photomultiplier tube and preamplifier. The output from one crystal was fed to a single channel analyzer. The energy of the gamma rays recorded by this analyzer and the energy width of the "window" of this analyzer were both adjustable. The pulse from the second preamplifier was cabled to a linear amplifier. The signal pulse from this amplifier and that from the single channel analyzer were lead into a coincidence circuit, the signal pulse from the single channel branch serving as the gate pulse. Only those signal pulses which were in coincidence with the gate pulses within the 10^{-6} second resolving time were passed on to the differentiating circuit of the 50 channel analyzer. Hence the gamma spectrum in coincidence with a particular gamma ray could be examined.

The chance rate was determined by placing an absorber between the sample and the gate crystal and applying a different radioactive source of about the same counting rate to the gate crystal. The usual calculation of the chance spectrum was carried out.

IV. EXPERIMENTAL RESULTS

1. 49.8 and 80 kev gamma rays.--Figure 1 indicates the gamma spectrum of AcK in the 0-150 kev range. The energy scale was calibrated with the 60 kev gamma ray in the decay of Am^{241} and the 87 kev gamma radiation of Cd^{109}. This and many similar curves were taken through 943 mg/cm^2 beryllium absorber to eliminate interference from the beta particles. There are prominent photopeaks at ~15, 50 and 80 kev, all decaying with the half-life of AcK.

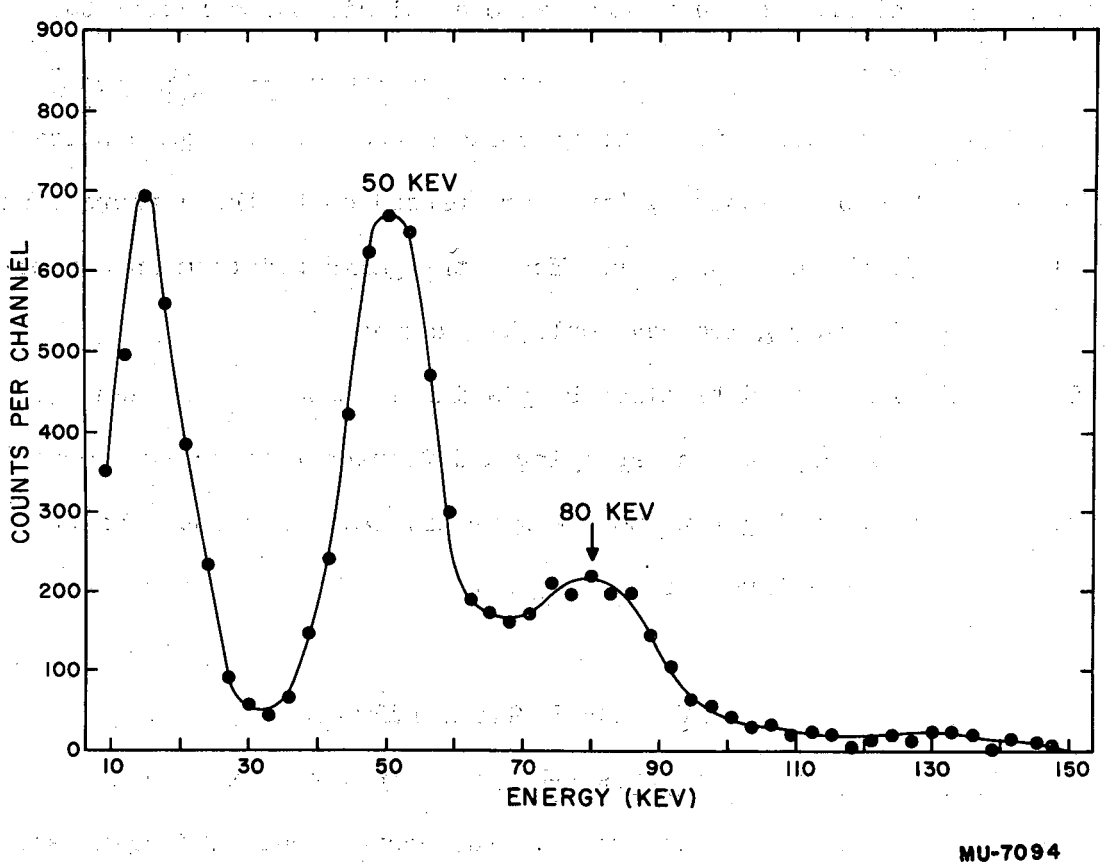

Figure 1. Gamma spectrum of AcK in the 0-150 kev range. Data taken through 943 mg/cm^2 beryllium absorber.

The ratio of the 80 kev/50 kev photopeaks was determined on several samples, the best value being 0.6 ± 0.1 The overlapping of the two peaks causes some uncertainty in this ratio.

The ratio of 50 and 80 kev photons to AcK disintegrations was determined in the following way. A sample of Am^{241} with a known alpha disintegration rate mounted on platinum disc was placed in the scintillation spectrometer on the same shelf position used for the AcK samples and counted through the 943 mg/cm^2 beryllium absorber. The area under the 60 kev photopeak was determined. Using the information that 40 of the 60 kev photons are emitted per 100 alpha disintegrations of Am^{241} the overall counting efficiency was determined as 5.4 percent, i.e., 5.4 percent of the 60 kev photons emitted by the sample are registered in the photopeak.

The absolute disintegration rate of AcK samples mounted on platinum was determined by counting the samples in the geiger counter and applying the calibration factor of 42 as discussed above. The counting efficiency of the 50 and 80 kev photon peaks was taken as 5.4 percent. The resulting absolute abundances were: 40 ± 10 of the 50 kev photons per 100 disintegrations of AcK and 24 of the 80 kev photons per 100 disintegrations. This calculation assumes that the variation in the ratio of the escape peak to the photopeak in the 50-80 kev range as a function of energy is negligible. Actually this is not true but unpublished experimental results of F. Asaro using the identical apparatus and geometrical conditions indicate that the variation is not great. Asaro obtained a ratio of the observed escape peak to the observed photopeak of 20 percent at 50 kev dropping to 16 percent in the region 60-100 kev. Hence the corrections to the above quoted abundances are only a few percent at most.

2. 215 and 310 kev gamma rays.--Figure 2 shows the gamma spectrum from 0-800 kev. It is to be noted that some gamma radiation appears in the region of 215 kev in low abundance compared to the 50 and 80 kev radiation. There is no other radiation with abundance >1 percent with respect to these peaks and additional curves showed this to be true through the 2 Mev range. The gamma radiation in the region of 215 kev was studied more carefully as shown in Figure 3, after calibrating the energy scale with annihilation radiation from a Na^{22} sample and with the 662 kev gamma radiation of Cs^{137}. A definite photopeak at 215 kev and a smaller peak at 310 kev were observed. The decay of these peaks was followed on numerous separately prepared samples to establish the 21-minute half-life. The ratios of these photopeaks to the 50 kev photopeak was determined correcting for the fact that the photoelectric efficiency of the crystal was not 100 percent for the 215 and 310 kev radiation. The absolute abundances were determined as 3-4 photons of 215 kev energy per 100 disintegrations and about 0.8 photon of 310 kev energy per 100 disintegrations of AcK.

3. Beta spectrum.--Several determinations of the maximum energy of the beta particles emitted by pure samples of AcK were made using the anthracene crystal. A visual endpoint energy was 1.15 ± 0.05 Mev. This confirms the 1.2 ± 0.1 Mev value previously obtained by Perey and Lecoin[7] by a measurement of tracks in a low pressure cloud chamber. The spectrum was not analyzed carefully for complexity.

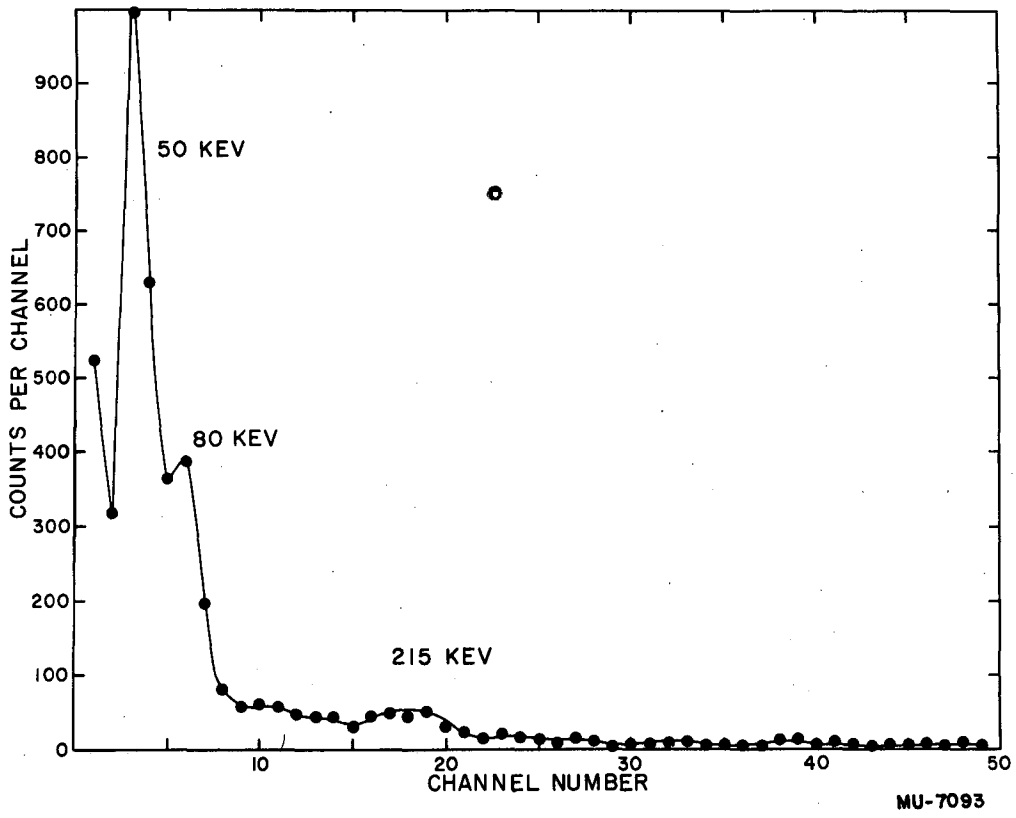

Figure 2. Gamma spectrum of AcK in the 0-800 kev region. Data taken through 943 mg/cm^2 beryllium absorber.

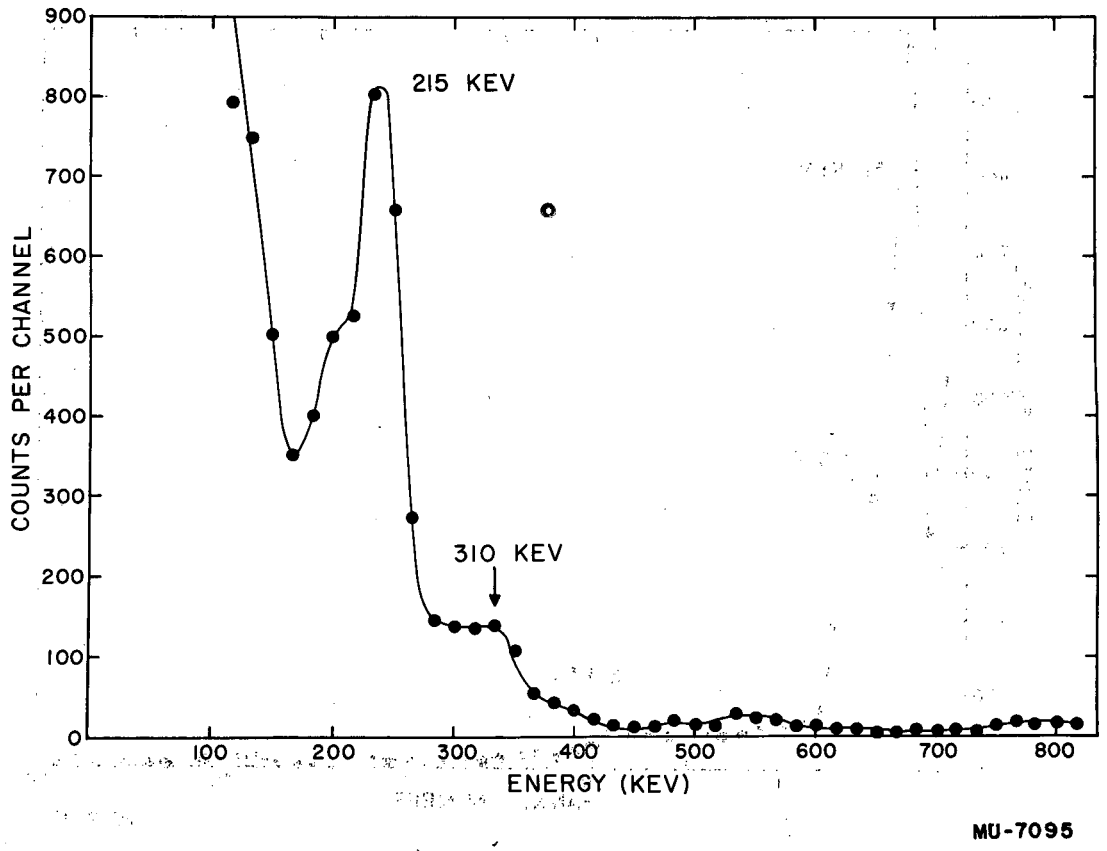

Figure 3. Gamma curve showing 215 kev and 310 kev radiation in AcK sample. Data taken through 943 mg/cm^2 beryllium absorber.

4. Proportional counter measurements on x-rays and the 50 kev gamma ray. Figure 4 shows the gamma spectrum of AcK in the low energy region taken with the proportional counter equipment. The data were taken with 706 mg/cm^2 beryllium absorber above the sample to remove beta particles. On the same figure are shown calibration curves taken on samples of Am^{241} and Io^{230}. In the Am^{241} curve are observed the prominent L_{α_1} (13.94 kev) and L_{β_1} (17.74 kev) lines of neptunium, a 26 kev gamma peak and an "escape" peak at 30 kev caused by the photoelectric ejection of a K electron from a xenon atom in the counter by the 59.8 kev gamma ray followed by the escape of the K_α x-ray of xenon from the tube. The ionium curve shows the prominent L_{α_1} (12.39 kev) and L_{β_1} (15.23 kev) lines of radium. It can be noted that AcK gives rise to identical radium L x-rays as well as to an escape peak at 20 kev resulting from the 50 kev gamma ray.

That this identification of the escape peak is correct was proved by redetermining the curve with a 1 gm/cm^2 aluminum absorber over the sample. The transmission of the x-rays through this thickness of aluminum is <0.1 percent while the transmission of the 50 kev gamma radiation is about 70 percent. A typical curve is shown in Figure 5. The energy scale was calibrated with Am^{241} and Io^{230} as before. To be noted is the appearance of a double escape peak attributable to the escape of K_{α_1} and K_{β_1} xenon x-radiation after absorption of the 50 kev gamma ray. These escape peaks serve as a better energy calibration than the sodium iodide crystal determinations of this value for the 50 kev gamma ray. The average of several determinations is 49.8 ± 0.3 kev.

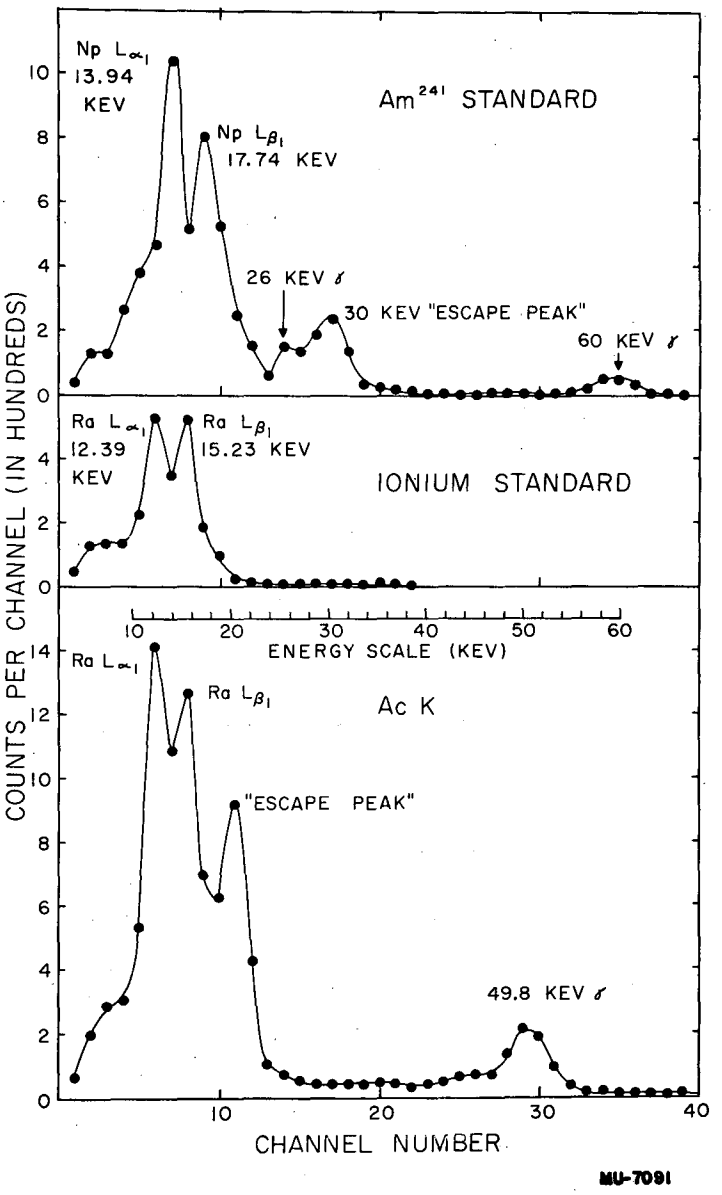

Figure 4. X-ray and soft gamma spectrum of AcK taken with proportional counter connected to 50 channel pulse height analyzer. Calibration runs on Am^{241} and ionium are shown. The AcK sample was mounted on aluminum. The 706 mg/cm^2 beryllium absorber was placed above sample to absorb β^- particles.

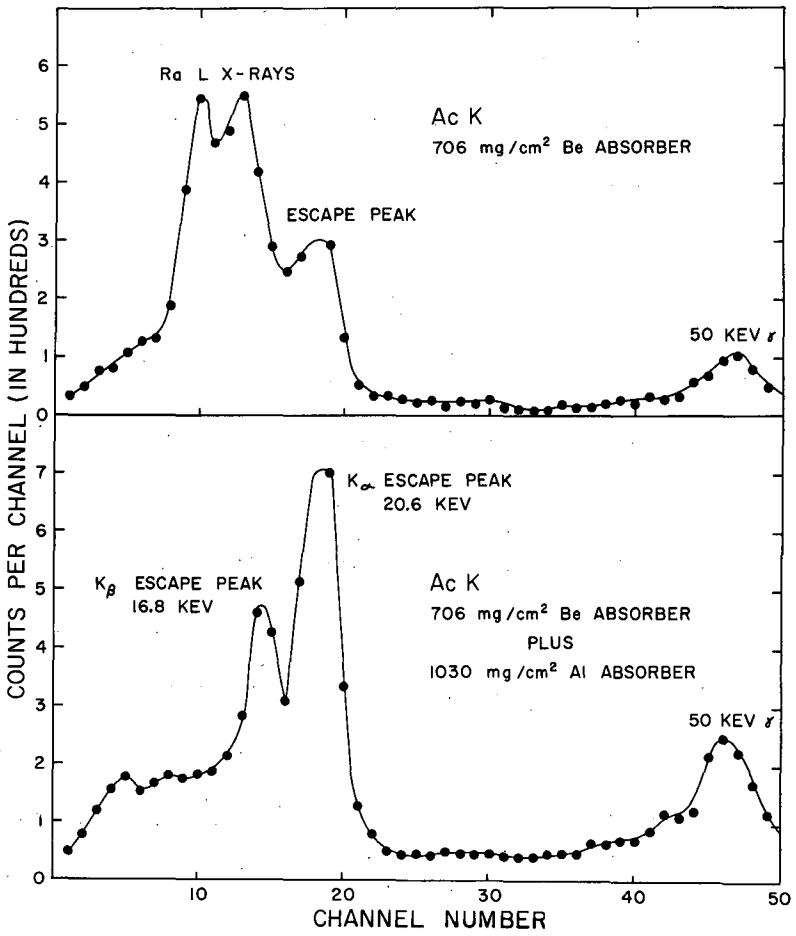

Figure 5. X-ray and soft gamma spectrum of AcK taken with proportional counter connected to 50 channel pulse height analyzer. Lower curve taken through 1032 mg/cm^2 aluminum absorber to absorb L x-rays. Resulting peaks are the escape peaks of the 49.8 kev gamma ray with escape of the K_α or K_β radiation of xenon. Data were taken for 1 minute for upper curve and for 4 minutes for lower curve.

5. Coincidence studies on AcK.--AcK samples mounted on quarter mil aluminum foils were placed between the two sodium iodide crystals of the coincidence apparatus and gamma-gamma coincidence spectra were run, gating in turn with the 50, 80 and 215 kev radiation. These measurements were performed by Mr. Frank Stephens. Some small coincidence peaks were observed but the order of intensity of these was very low. The 50-80 kev gamma-gamma coincidence peak was less than 1/100 of that to be expected if these rays were in coincidence. The 215-50 kev and 215-80 kev coincidences, with the 215 kev serving as the gate, were less than 1/30th of those to be expected of a true gamma-gamma coincidence. Of considerable importance to the consideration of various possible decay schemes is the presence or absence of a 50-30 kev coincidence or a 50 kev L x-ray coincidence, the L x-rays coming from the conversion of a 30 kev gamma ray. The absorption of the soft x-rays in the crystal cover and the fluorescent yield reduce the sensitivity of this determination considerably but the intensity was conservatively a factor of 5 too low to indicate a 50 kev L x-ray coincidence and within considerable larger limits there was no sign of a 50-30 kev gamma-gamma coincidence. The significance of this is that the 50 and 80 kev transitions must both lead to the ground state of Ra^{223}.

6. Study of radiations of Th^{227}.--Since Th^{227} (RdAc) and AcK decay to the same daughter product it might be expected that some Ra^{223} levels would be common to the decay schemes of both isotopes. In particular, levels with the 50 and 80 kev spacing of the two most prominent gamma rays in the decay of AcK might be expected among the lower lying

levels of Ra^{223}. The alpha spectrum of Th^{227} is exceedingly complicated with 11 reported groups of greater than 1 percent abundance. Figure 6 displays the results of a study of the alpha groups carried out by Rosenblum et al.[8] The Ra^{223} levels are labeled in kev above the ground state using the assumption that the most energetic alpha particle leads to the ground state, an assumption which is probably false since alpha decay to the ground state of an odd nucleon isotope is frequently more hindered than that to a higher lying level.[9]

There are several literature reports on the gamma rays of Th^{227}. The absorption curve studies of Riou[10] showed a 50 kev gamma ray in 3 percent abundance, a 126 kev gamma ray in 13 percent abundance, and a 280 kev gamma ray in 50 percent abundance. Similar results were reported by Bachelet and Savel.[11] Studies on a crystal spectrometer were reported somewhat earlier by Frilley[12] who found gamma rays at 50, 57, 80, 101, 113, 119, 208, 240, and 258 kev. Magnetic analysis of the electron spectrum by Surugue[13] and Te-Tschao and Surugue[14] showed 30 gamma rays between 30 and 638 kev.

Figures 7 and 8 indicate the gamma spectrum of a freshly purified sample of Th^{227} as obtained in our sodium iodide crystal spectrometer. The sample was purified by extracting the thorium isotope from a dilute acid solution of Ac^{227} into undiluted tributyl phosphate and washing the solvent repeatedly with 4 M nitric acid to remove possible radioactive contaminants. The most prominent photopeaks appear at 50, 87 and 235 kev. The last energy was determined to ±5 kev by calibration with the 662 kev gamma ray in the decay of Cs^{137}, annihilation radiation, the 59.6 kev gamma ray of Am^{241} and the 184 kev gamma ray of U^{235}. The

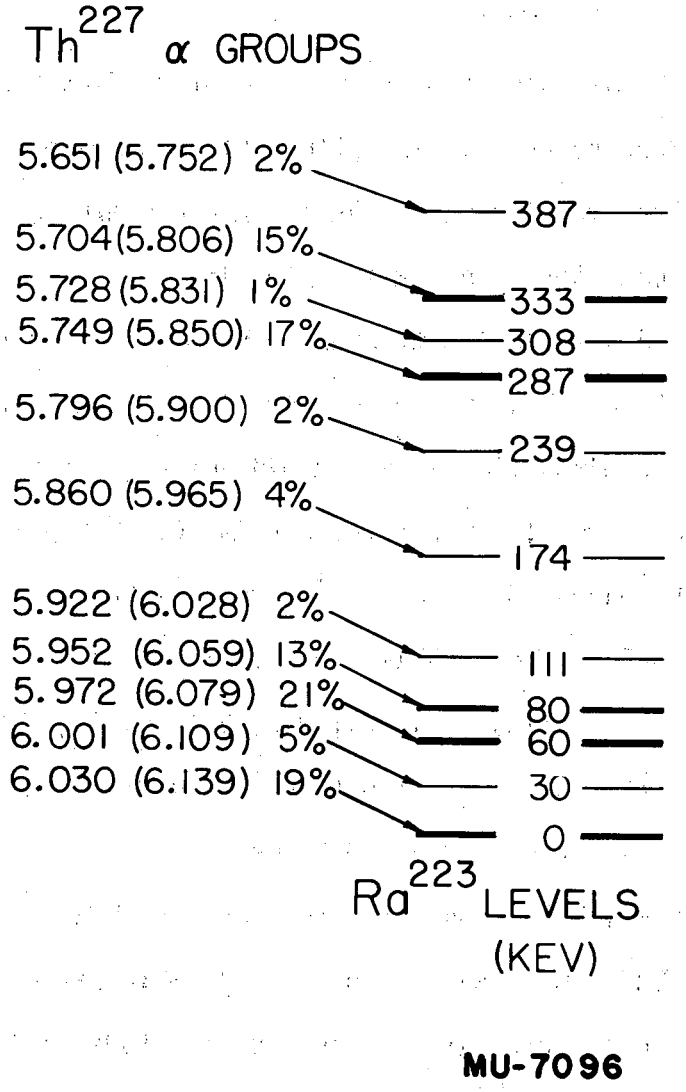

Figure 6. Ra^{223} levels reached by alpha decay of Th^{227}. Alpha particle energies (in Mev) and abundances as reported by Rosenblum et al.[8] are shown. Figures in parentheses are alpha disintegration energies (i.e., the recoil energy has been added to the alpha particle energy). The accentuated Ra^{223} levels are those which are reached by alpha groups greater than 10 percent in abundance.

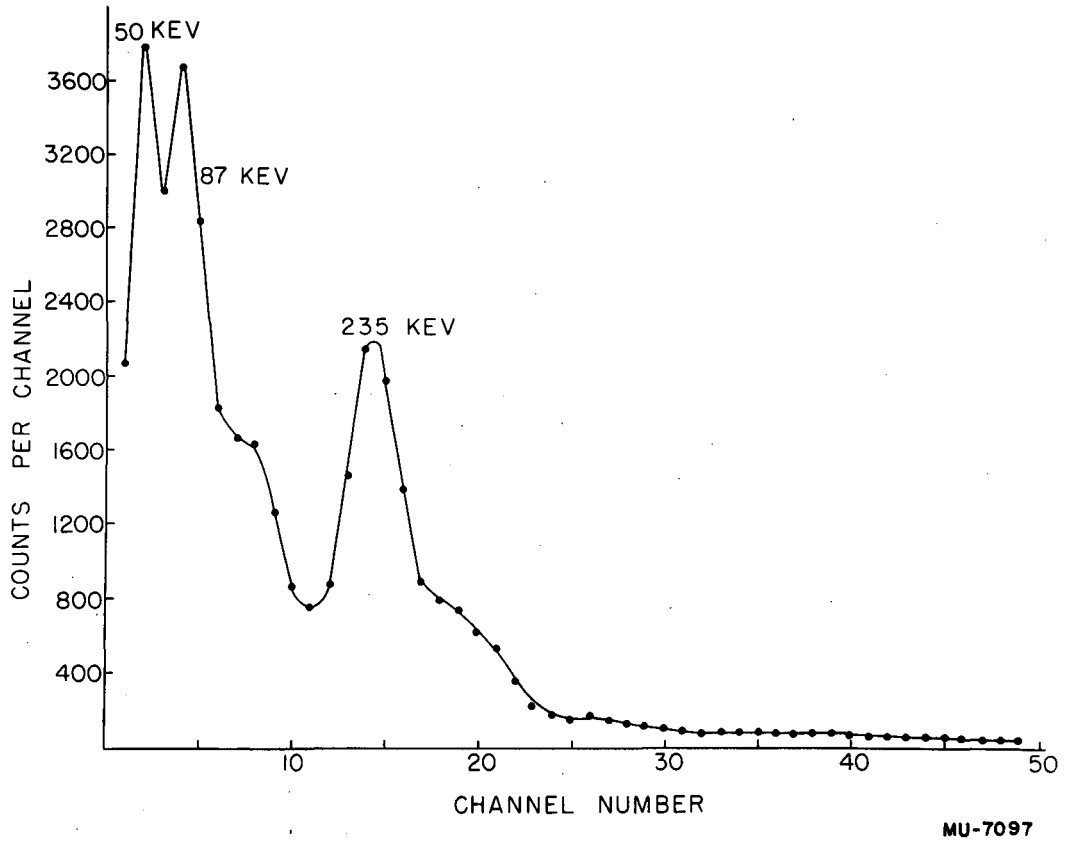

Figure 7. Gamma spectrum of Th^{227} as determined on sodium iodide crystal spectrometer in 0-800 kev region.

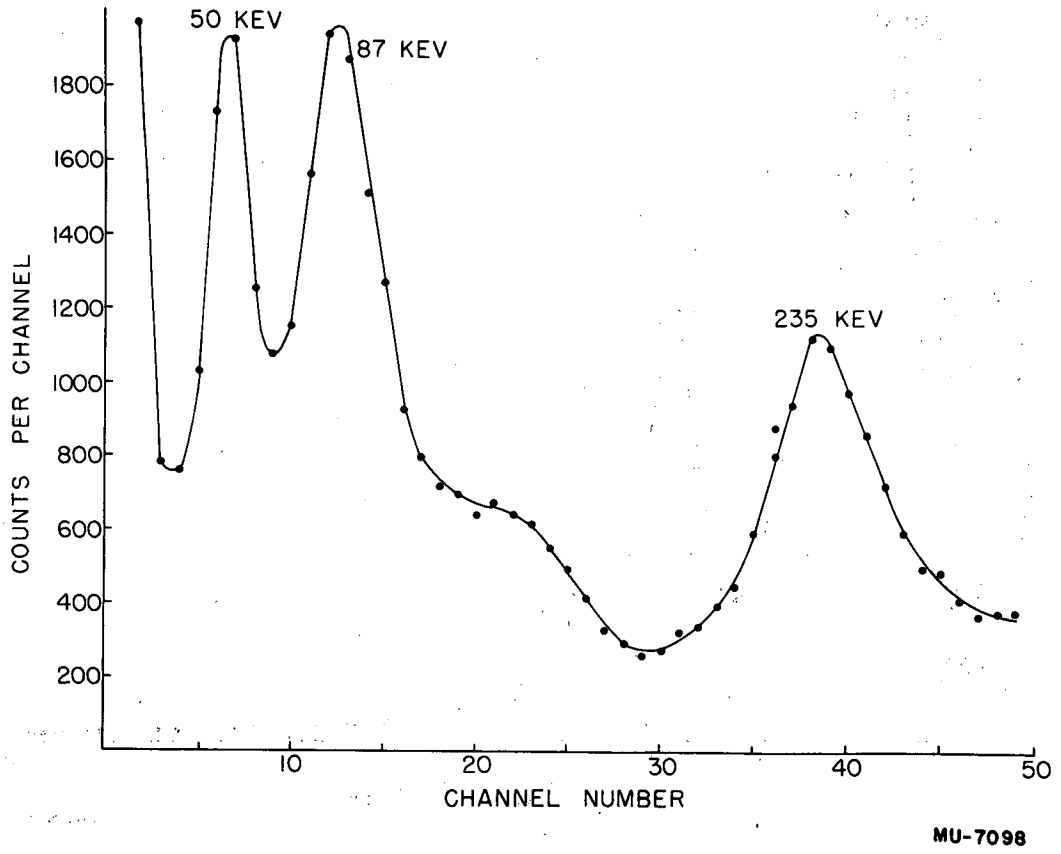

Figure 8. Gamma spectrum of Th^{227} in 0-300 kev region.

absolute abundance of the 50 kev gamma ray was determined roughly to be 0.2 photons per total alpha particle. Gamma-gamma coincidence studies were carried out for us by Mr. Frank Stephens with the equipment described previously, gating in turn with the 50 and 235 kev gamma rays. Figure 9 shows a prominent coincidence peak between the 50 and 235 kev radiations. This coincidence is so prominent and so easily reproduced that it has been found to be quite useful in our laboratory as a standard for other gamma-gamma coincidence studies. In the run with the 50 kev gate some coincidence peaks at ~90 and ~140 kev of much lower intensity (~1/20) were observed.

It seems likely that the 50 kev gamma ray is identical with the 49.8 kev gamma ray of AcK and this belief was strengthened by determining the energy of the gamma ray in the proportional counter. The K_{α_1} and K_{β_1} escape peaks were determined as described above for AcK and as illustrated in Figure 5. Freshly purified samples of Th^{227} and of AcK were counted through 1 gm/cm^2 of aluminum absorber within a few minutes of each other to eliminate any errors due to a drift in the calibration of the proportional counter. The curves of the escape peak were both identical with Figure 5. It is believed that a difference of 1 kev could easily have been detected.

V. DISCUSSION

A detailed study of the radiations of Ac^{227} and AcK has previously been carried out by Lecoin and co-workers[2] using xenon filled geiger counters and absorption curves. Their findings on AcK were as follows. A prominent gamma ray of 48.6 kev energy is emitted in high abundance

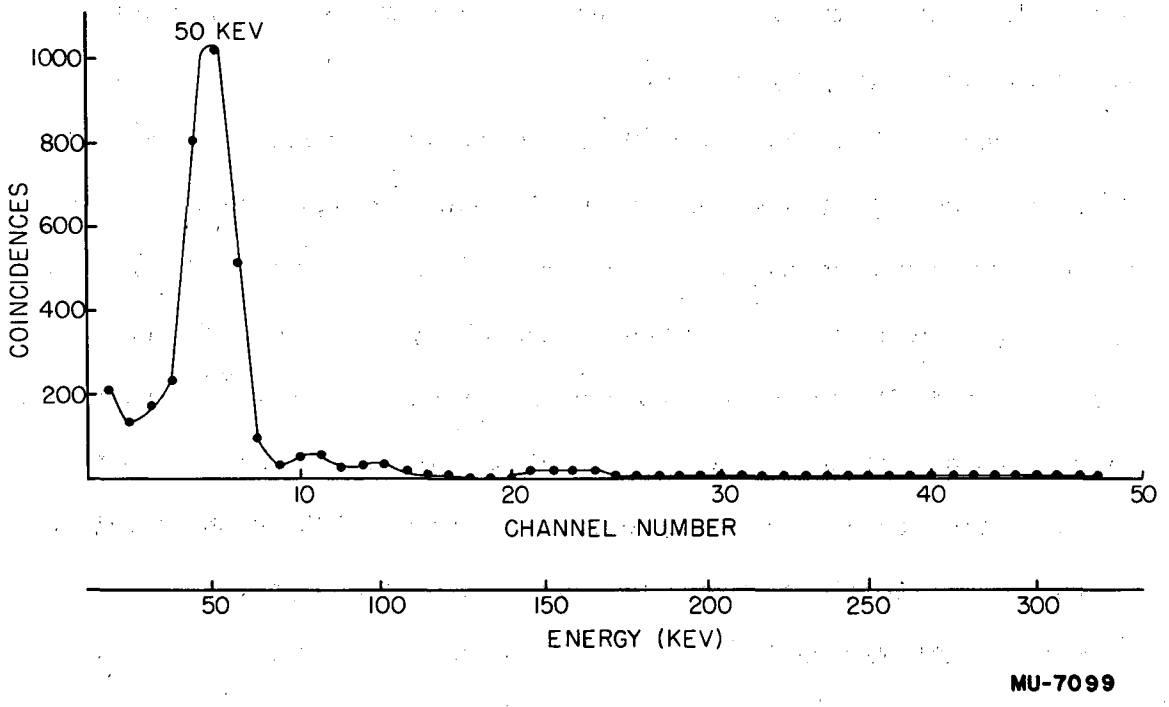

Figure 9. Gamma spectrum in coincidence with 235 kev gamma ray of Th^{227}.

(27 photons per 100 disintegrations). L radiation of 15 kev is emitted in 25 disintegrations out of 100. A gamma ray of 330 kev is emitted at the rate of 6 photons per 100 disintegrations.

The present results are in good agreement with respect to the 50 kev radiation. Lecoin and co-workers determined a precise value of 48.6 ± 1.6 kev by the method of critical absorption. Various elements between Z = 53 and Z = 74 were used as absorbers and a marked change in absorption between $_{63}Eu$ and $_{64}Gd$ bracketed the gamma energy. The quoted value could be raised somewhat as the recent Table of Critical X-Ray Absorption Energies of Hill et al.[15] give 48.51 and 50.23 kev for the K shell energies of europium and gadolinium. Our best value determined from the escape peak values as mentioned above is 49.8 ± 0.3 kev.

The French authors also bracketed the 14.5 kev radiation by the method of critical absorption using $_{33}As$, $_{34}Se$ and $_{38}Sr$ absorbers, and showed that it was the L radiation of radium.

For radiation of energy greater than 50 kev the agreement between the two studies is not as good. The prominent radiation of 80-85 kev is not reported by Lecoin and co-workers.[2] The gamma ray of 330 kev reported by these authors with an intensity of 6 photons per 100 disintegrations may perhaps be identified with the 215 kev radiation with an intensity of 3-4 photons per 100 disintegrations, reported here since energy determinations by absorption measurements have a tendency to give high values.

It may be mentioned that the 80 kev radiation reported here is not K x-radiation since the K_{α_1} and K_{α_2} lines of radium would be expected

at 88.5 and 85.4 kev, although it is quite possible that some K radiation is included in the high energy side of the 80 kev peak.

It was hoped that the gamma rays of AcK could be fitted into a decay scheme using the Ra^{223} levels revealed by alpha spectrum analysis of RdAc (Figure 6) and consistent with the gamma radiation of Th^{227}. The chief facts which must be taken into account are: 1) The 49.8 and 80 kev gamma rays observed in the decay of AcK connect low-lying levels in Ra^{223}; 2) The 49.8 kev gamma ray is not in coincidence with 80 kev radiation, 30 kev gamma radiation or L x-radiation; 3) The same 49.8 kev gamma transition is observed in the decay of AcK and RdAc (Th^{227}); 4) In the decay of RdAc the 49.8 kev gamma ray is in coincidence with a 235 kev gamma ray.

It was impossible to construct a partial decay scheme consistent with these data using only the levels of Figure 6. It is necessary to postulate other low-lying levels and it is quite probable that the ground state of Ra^{223} lies below the level so labeled on Figure 6. This is not unreasonable in view of the well established fact that alpha decay to the ground state is usually strongly hindered for alpha emitters with odd nucleons.[9] Further work to give more definite information on the low-lying levels of Ra^{223} is contemplated.

ACKNOWLEDGMENTS

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