gested several of the chemical separations. Thanks are also due Mr. Bernie Rossi and the 60-inch cyclotron crew for carrying out the bombardments.

APPENDIX. CHEMICAL PROCEDURE

To the solution of Mn in HCl 10 mg amounts of Co, Cu, Fe, Ni, Zn carriers were added. The 1*N* solution was saturated with $(NH_4)_3SCN$ and shaken with a 50 percent mixture of diethyl ether and amyl alcohol. After several washings the alcohol-ether extract was found to be free of Mn. The remaining metals were removed from the extract by shaking with an ammoniacal solution, the copper precipitated as sulfide in 0.3*N* acid solution, the nickel separated as the dimethyl glyoxime, the zinc obtained as sulfide in nearly neutral solution, the iron precipitated as the hydroxide and finally the cobalt separated as sulfide in a basic solution or precipitated by nitroso-β-naphthol. This procedure in parts or as a whole was repeated several times. After the assignment to the element had been established, 1 mg amounts of Co carrier were extracted from a solution of the manganese target with the alcohol-ether mixture, and directly precipitated as the sulfide from an ammoniacal solution.

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Decay Constants of K^{40}

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New measurements have been made of the decay constants of K^{40} for beta-ray emission and for electron capture, and a decay scheme is proposed on the basis of these measurements and the previous data. Special counters adapted to the measurement of low levels of activity were developed for making the measurements. The results obtained were 28.3 ± 1.0 beta-rays per gram of K per sec. and 13.5 ± 4.0 electron captures per 100 beta-rays, corresponding to a total half-life of $12.7\pm0.5\times10^8$ yr., with 88 percent of the decays by beta-ray emission and 12 percent by electron capture. A previous measurement of the gamma-ray intensity in K^{40}, by the present authors, gave 3.6 ± 0.3 gamma-rays per gram of K per sec., or about 12.7 gamma-rays per 100 beta-rays. Since the numbers of gamma-rays and of electron captures in the decay of K^{40} are essentially the same, and since previous measurements indicate that the gamma-energy is greater than the beta-energy, it is proposed that the gamma-ray be associated with the electron-capture decay, a gamma-ray following each electron capture.

I. INTRODUCTION

POTASSIUM 40 is known to decay by beta- and gamma-ray emission and by electron capture, but recent measurements of the decay constants have been in rather poor agreement. The most reliable recent measurements[1–6] of the beta-half-life have given values ranging from 12.0 to 17.8×10^8 yr., with an average value of about 14.4×10^8 yr. The most ingenious method was that of Stout,[4] who mixed into his potassium sources an accurately known activity of Na^{24}, which has nearly the same beta-energy as K^{40}, and measured the ratio of K^{40} to Na^{24} counting rates. Knowing the specific activity of the Na^{24}, he was then able to determine the K^{40} activity directly, without corrections, and obtained 13.1×10^8 yr.

There is even more disparity between the recent estimates of electron-capture intensity, the values ranging from 5 to 190 electron captures per 100 beta-rays. The discrepancies here can be attributed to the uncertainties inherent in the methods used in making the measurements. The measurement of Bleuler and Gabriel[7] was made with a counter arranged to admit gaseous absorbers to absorb selectively the x-rays resulting from the electron-capture decay. Most of the counting rate was due to beta-rays from K^{40}, and the reduction in counting rate when the absorber was introduced was only about two percent. Scattering of the beta-rays in the gaseous absorber may have caused a large part of the two percent change in their counting rate, and the result of 190 electron captures per 100 beta-rays is probably considerably too large. The other determinations[2,8,9] of electron-capture intensity were made from the amount of argon trapped in potassium-bearing minerals, on the assumption that the argon had been produced by decaying potassium. The most refined of these measurements was that of Aldrich and Nier,[10] who used the mass spectrograph to analyze the ratio of A^{40} to A^{36} in minerals. They found that A^{40} was relatively more abundant in potassium-bearing minerals than in present-day atmospheric argon, and from their data they estimated that there are about 10 electron captures per 100 beta-rays in K^{40}.

New measurements of the decay constants for beta-ray emission and for electron capture were undertaken.

[1] T. Gráf, Phys. Rev. **74**, 831 (1948).
[2] L. B. Borst and J. J. Floyd, Phys. Rev. **74**, 989 (1948).
[3] O. Hirzel and H. Wäffler, Phys. Rev. **74**, 1553 (1948).
[4] R. W. Stout, Phys. Rev. **75**, 1107 (1949).
[5] J. J. Floyd and L. B. Borst, Phys. Rev. **75**, 1106 (1949)
[6] F. W. Spiers, Nature **165**, 356 (1950).
[7] E. Bleuler and M. Gabriel, Helv. Phys. Acta **20**, 67 (1947).
[8] L. H. Ahrens and R. D. Evans, Phys. Rev. **74**, 279 (1948).
[9] H. E. Suess, Phys. Rev. **73**, 1209 (1948).
[10] L. T. Aldrich and A. O. Nier, Phys. Rev. **74**, 876 (1948).

A measurement of the gamma-ray intensity, yielding a value of 3.6±0.3 gamma-rays per gram of K per sec. has already been reported.[11]

II. BETA-RAY MEASUREMENTS

A. The Beta-Counter

Measuring the beta-activity of K^{40} is difficult because the activities of available sources are weak, and sources of large area must be used to achieve measurable counting rates. Obviously, the best possible arrangement for measuring the weak activity of K^{40} would be one in which a source of large area could be placed inside a counter in such a manner that every beta-ray leaving the source would be counted. To this end, a special counter was constructed (Fig. 1), using an adaptation of the cell counters developed by Beyster and Wiedenbeck.[12] The counter uses two independent, pancake-like cell-sections. Basically, each section is a short brass cylinder, 6 in. in diameter and $1\frac{1}{4}$ in. high, closed at both ends. The anode wire of each section is a 10-mil tungsten wire, bent into a ring 3 in. in diameter and supported in the center of the section by glass rods which are sealed through the wall of the cylinder. The two cells are placed together, with a common cathode surface between them, and operated as a single counter. This common cathode surface is a thin aluminum-foil disk on which the source of K^{40} is mounted. The counter is so arranged that, for background measurements, the source-bearing foil can be replaced by a blank foil without breaking the vacuum seal of the counter. The two aluminum foils, one blank, and the other bearing the source, are mounted on a tray which can be moved back and forth by a rack-and-pinion arrangement. Thus either foil can be centered in the counter while the other foil moves out into one of the wings of the counter.

This beta-counter, because of the geometrical arrangement, has the advantages over conventional counters that sources of considerable area can be used (about 120 cm^2) and that every beta-ray which leaves the source enters the sensitive volume of the counter.

B. Performance of the Beta-Counter

Before a counter of such unconventional design could be used to measure the absolute number of disintegrations in a sample, it was necessary to prove that it had the proper characteristics, namely, that it actually counted every beta-ray leaving the source and that it added no additional spurious counts itself. Accordingly, tests were made of its plateau, sensitivity, and efficiency.

The counter was filled to 10 cm pressure with a mixture of 90 percent argon and 10 percent alcohol.

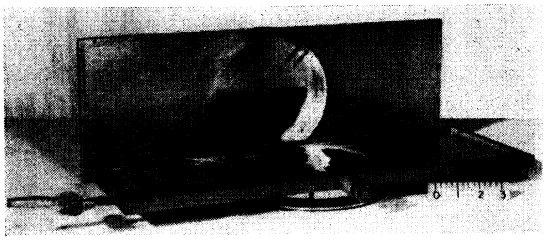

FIG. 1. View of the beta-counter disassembled.

Although the counter was self-quenching, an auxiliary quench circuit was used, because with it the plateau was flattened considerably. The plateau of the counter was about 125 volts long, and the rise in counting rate along it was about three percent per 100 volts.

The sensitivity of the counter was tested by placing a point souce of Tl^{204}, a pure beta-emitter, on the sliding tray inside the counter and measuring the counting rate as the source was moved in $\frac{1}{4}$-in. steps across the diameter. The counter was found to be uniformly sensitive except near the walls and in the exact center, where a small dip was observed in the sensitivity. This dip is not surprising, because a particle leaving the source at the center and passing vertically upward through the counter travels through a region of zero field. A correction of two percent was made to the measured activity of K^{40} to allow for the lack of uniform sensitivity.

The efficiency of the counter was measured by the coincidence method and was found to be greater than 98 percent. Calculations based on the known specific ionization caused in argon by beta-particles[13] indicated that the efficiency of the counter was about 99 percent. Accordingly, a one percent correction was made to the K^{40} data for counter inefficiency.

The tests just described demonstrated that the counter had a flat plateau, high efficiency, and nearly uniform sensitivity and that it was, therefore, suitable for use in measuring the activity of K^{40}.

C. Measurements with Ordinary Potassium

For the measurements on ordinary potassium, sources of chemically pure KCl were used. A saturated solution of KCl in water was sprayed onto aluminum-foil blanks in a fine mist. Any desired thickness could be deposited; the amount deposited was determined by accurate weighings of the blank, both before and after spraying.

Twenty sources were made, of thickness varying from 0.5 to 13 mg/cm^2, and on three weights of aluminum-foil backing: 0.18, 2.67, and 5.43 mg/cm^2. Each of these sources was placed in the counter, together with a blank foil identical with the source-carrying foil, and counting runs were made, alternating source and background

[11] G. A. Sawyer and M. L. Wiedenbeck, Phys. Rev. **76**, 1535 (1949).

[12] J. R. Beyster and M. L. Wiedenbeck, Rev. Sci. Inst. **19**, 819 (1948).

[13] S. A. Korff, *Electron and Nuclear Counters* (D. Van Nostrand Company, Inc., New York, 1946), p. 70.

measurements every 1000 sec. It was found in practice that if the sources were not covered with an aluminum foil, the counting-rate in the counter section faced by the open source increased until the counter went into a continuous discharge. The cause, apparently, was an electrical charging of the insulating surface of KCl. Accordingly, all sources were covered with aluminum foil of the same weight as the backing foil.

From the data obtained with various source- and backing-thicknesses, it was possible to extrapolate the measured activity of the K^{40} beta-rays to zero source-thickness and zero backing-thickness, and this extrapolated value was found to be 28.3 disintegrations per gram of K per sec. This specific activity corresponds to a half-life of 14.2×10^8 yr. for beta-decay in K^{40}, using the recent value[14] of Nier, 0.0119 ± 0.0001 percent, as the isotopic abundance of K^{40} in ordinary potassium.

The statistical counting errors in the experiment are less than one percent except for the thinnest sources. An indication that the statistical errors are small is the fact that in extrapolating the activity to zero source-thickness, when a straight line was drawn through the plotted data, only one point fell more than one percent off the line. Systematic errors, such as those caused by counter inefficiency, are probably not more than two or three percent. The value for the half-life, therefore, is given as $14.2\pm0.5\times10^8$ yr.

D. Measurements with an Enriched Source

A small sample of KCl containing K^{40} enriched to isotopic abundance 0.40 ± 0.02 percent was obtained from the Carbide and Carbon Chemicals Corporation, Oak Ridge, Tennessee, and a source was made from this sample by evaporating the KCl from a tungsten filament onto a silvered zapon film. Source- and backing-thickness were both about 0.1 mg/cm². A measurement of the beta-half-life was made with this source, in the special beta-counter. The measured half-life was $13.0\pm0.7\times10^8$ yr., the largest contribution to the error being in the isotopic abundance of K^{40} in the source. This value barely agrees, within the probable errors, with the measurements made with ordinary potassium. Since only a single measurement was made and the uncertainties are greater, this value is less reliable than the measurements with ordinary KCl sources and, therefore, was not used; the half-life was taken as 14.2×10^8 yr., as measured with the ordinary KCl sources.

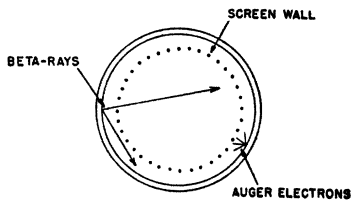

FIG. 2. End view of the electron-capture counter showing paths of beta-rays and Auger electrons.

[14] A. O. Nier, Phys. Rev. **77**, 789 (1950).

III. ELECTRON-CAPTURE MEASUREMENTS

A. The Electron-Capture Counter

The electron-capture decay in K^{40} can be positively identified through the characteristic x-rays emitted by the daughter nucleus, A^{40}, as the vacancy in its K shell is refilled. However, in the case of argon, 88 percent of the x-rays are converted to Auger electrons of 2.9-kev energy,[15] and these Auger electrons can be used, instead, as a measure of the electron capture.

The chief problem in measuring the electron-capture intensity with a Geiger counter is to distinguish between the Auger-electron counts and the beta-ray counts which also come from K^{40}. To investigate this problem, a screen-wall counter was constructed. The counter is contained in a cylindrical steel shell, $1\frac{1}{4}$ in. in diameter and 6 in. long. The counter cathode is a cylindrical screen, 1 in. in diameter, placed inside the steel shell and insulated from it, while the anode of the counter is a 12-mil tungsten wire placed axially in the center of the counter. The special screen-cathode was made by winding a "squirrel-cage," or cylindrical grid, of 10-mil copper wire, on a frame consisting of two copper rings, 1 in. in diameter, spaced 5 in. apart and supported by tungsten rods sealed through the end plate of the steel shell. A small bias voltage can be applied between the screen and the shell of the counter. The radioactive source was mounted in a thin layer on the inside surface of the counter shell.

This counter arrangement permitted the Auger electrons to be distinguished from the beta-rays in the counter. The range of the Auger electrons in the counter gas is only a few millimeters and so they were stopped between the shell of the counter and the screen cathode and did not get into the sensitive region of the counter; the beta-rays from the source, on the other hand, are much more energetic, and they were able to pass through the screen into the sensitive region of the counter to be counted. Now, if a bias voltage is applied between the screen and the counter shell so that the screen is negative, the secondary electrons produced by the Auger electrons in ionizing the counter gas between the shell and the screen will be drawn to the shell of the counter and will not be counted. But if the bias voltage is reversed, so that the screen is positive with respect to the shell, the secondary electrons produced by the Auger electrons are drawn through the screen into the sensitive region of the counter and are therefore counted.

The small bias voltage applied (30 volts) has no effect on the trajectory of the high energy beta-rays, and one might, at first thought, expect the beta-ray

[15] A. H. Compton and S. K. Allison, *X-Rays in Theory and Experiment* (D. Van Nostrand Company, Inc., New York, 1935), p. 488.

counting rate to be independent of it. However, applying the positive bias to the screen does increase the number of beta-ray counts, because some of the beta-rays leave the source in such a direction that they do not pass through the screen into the counting region, as can be readily seen from the end view of the counter in Fig. 2. When the screen is made positive, the secondary electrons produced by these beta-rays are drawn through it and are counted, while with the screen negative, they are not counted. Thus, the beta-rays which do not enter the sensitive part of the counter produce exactly the same effect as the Auger electrons, and they must, therefore, be taken into account.

B. Performance of the Counter

Figure 3 shows the counting rate of the counter as a function of bias voltage between shell and screen. It can be seen that at both positive and negative 30-volt bias—the bias voltage used in the measurements with the counter—the counting rate is independent of the bias. This fact indicates that with the screen at 30 volts negative, all secondary electrons formed between shell and screen are prevented from entering the sensitive region of the counter, and that with the screen at 30 volts positive, all secondary electrons formed between shell and screen are drawn into the sensitive region and are therefore counted.

Two beta-ray emitting sources, Na^{24} and K^{42}, were tried in the counter to determine exactly how much the counting rate for beta-rays changed as the screen-bias voltage was switched from negative to positive. The ratio of screen-positive to screen-negative counting rates for the two beta-ray sources was 71.9 and 70.0 percent, respectively. In other words, approximately 71 percent of the beta-rays pass into the sensitive part of the counter.

To determine the characteristics of the counter for Auger electrons, a source of Cr^{51} was used. Cr^{51} decays by electron capture,[16] with 97 percent of the disintegrations going directly to the ground state of V^{51}. The other three percent are by electron capture to excited states of the V nucleus, with emission of gamma-rays as the V falls to its ground state. In the case of V, about 75 percent of the x-rays are converted to Auger electrons,[15] a smaller number than in the case of A^{40} because V has a higher atomic number. The ratio of screen-negative to screen-positive counts for Cr^{51} in the counter was found to be seven percent—vastly different from the ratio for beta-rays. For pure Auger electrons, the ratio should be zero, if all the electrons are stopped in the gas before reaching the screen. The seven percent remaining here is attributed to the V x-rays emitted following 25 percent of the disintegrations.

To establish that all of the Auger electrons were stopped in the gas before reaching the screen, the ratio of screen-negative, to screen-positive counts was determined as a function of gas pressure in the counter. The counting rate with screen-positive was independent of pressure, as would be expected, but the counting rate with screen-negative increased if the gas pressure was reduced below 13 cm Hg. This increase was clearly caused by Auger electrons whose range, at the lower gas pressure, was sufficient to enable them to reach the screen and the sensitive counting region of the counter. The ratio of screen-negative to screen-positive counts as a function of gas pressure is shown in Fig. 4. It is clear that 13 cm of gas pressure is sufficient to stop the Auger electrons from V, which have an energy of 5 kev, and it is therefore adequate to stop the 2.9-kev Auger electrons from A. Accordingly, this was the gas pressure used in the experiments with the K^{40} source.

C. Measurements on K^{40}

For the determination of the electron capture in K^{40}, an extremely thin source of KCl, enriched in K^{40} to isotopic abundance 0.40 percent, was prepared by evaporating the KCl, in a vacuum, from a tungsten filament onto an aluminum-foil blank. The average thickness of this source was about 10 $\mu g/cm^2$, which was approximately one-sixth the range of the Auger electrons from A. The real counts from this source were about half the background counting rate. Several long runs were made with this source, alternated with

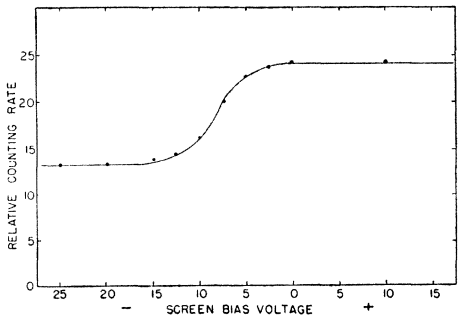

FIG. 3. Counting rate as a function of screen-bias voltage in the electron-capture counter.

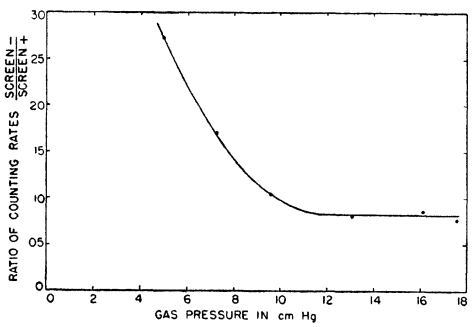

FIG. 4. Ratio of negative-screen to positive-screen counting rate as a function of gas pressure in the electron-capture counter.

[16] Bradt, Gugelot, Huber, Medicus, Preiswerk, and Scherrer, Helv. Phys. Acta 18, 259 (1945).

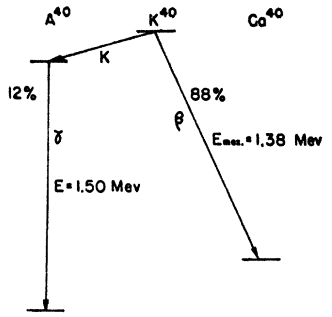

FIG. 5. Proposed decay scheme for K^{40}.

background runs, and a ratio of screen-negative to screen-positive counting rates of 64.4 percent was obtained. This ratio is to be compared with the values of 71.9 percent and 70.0 percent found for the two beta-ray emitters. It is clear that the ratio is slightly lower for K^{40} than for pure beta-ray emitters, a fact which indicates that Auger electrons are present. The number of Auger electrons calculated from these data is 12/100 beta-rays. Since, in addition, x-rays are emitted in 12 percent of the electron-capture decays and these are not detected, the actual number of electron captures is about 13.5/100 beta-rays.

Because of the uncertainty in the corrections for backscattering from the counter wall and for self-absorption in the source, the error in this value is fairly large, possibly as much as 30 percent, and it should not be regarded as exact. It does show, however, that electron capture actually takes place in K^{40}, and in about the same amount as the gamma-ray emission, there being, according to our previous measurement,[11] about 12.7 ± 1.1 gamma-ray per 100 beta-rays.

This result is consistent with the findings of Suess[9] and Aldrich and Nier.[10] The value of Bleuler and Gabriel[7] (190 electron captures per 100 beta-rays), however, is definitely ruled out, since such a large number of electron captures would mean a ratio of screen-negative to screen-positive counts of 30 percent in the present experiment.

III. CONCLUSIONS

When the decay constants of K^{40} for beta-decay (28.3 beta-rays per gram of K per sec.) and electron capture (13.5 electron captures per 100 beta-rays) are combined, the total half-life of K^{40} is calculated to be $12.7\pm0.6\times10^8$ yr.

A decay scheme for K^{40}, shown in Fig. 5, is proposed, which associates the gamma-ray with the electron-capture disintegration to A^{40}, rather than with the beta-decay to Ca^{40}. The reasons for this association are: (1) The most reliable measurements of the beta-ray[5,17–21] and gamma-ray[17,22–24] energies indicate that the gamma-ray energy is greater than the maximum beta-ray energy, and since there are many more beta-rays than gamma-rays in the decay of K^{40}, the gamma-ray cannot easily be fitted into a scheme placing it in cascade with a beta-ray. If the gamma-rays were associated with the beta-ray disintegrations, it would be necessary for part of the beta-decay to be to an excited state of Ca^{40}, followed by gamma-rays to the ground state of Ca^{40}, while the rest of the beta-decay would be directly to the ground state of Ca^{40}. These parallel processes are clearly impossible if no beta-rays have more energy than the gamma-rays. (2) The numbers of electron captures and gamma-ray emissions in K^{40} decay are equal, within the experimental errors, a fact which indicates that there is one gamma-ray for each electron-capture disintegration and that all electron-capture disintegrations are to an excited state of A^{40}, with a gamma-ray following.

[17] O. Hirzel and H. Wäffler, Helv. Phys. Acta **19**, 216 (1946).
[18] Dželepow, Kopjava, and Vorobjov, Phys. Rev. **69**, 538 (1946).
[19] W. J. Henderson, Phys. Rev. **71**, 323 (1947).
[20] D. Alburger, Phys. Rev. **75**, 1442 (1949).
[21] Bell, Weaver, and Cassidy, Phys. Rev. **77**, 399 (1950).
[22] E. Gleditsch and T. Graf, Phys. Rev. **72**, 640 (1947).
[23] P. R. Bell and J. M. Cassidy, Phys. Rev. **77**, 409 (1950).
[24] Pringle, Standil, and Roulston, Phys. Rev. **77**, 841 (1950).

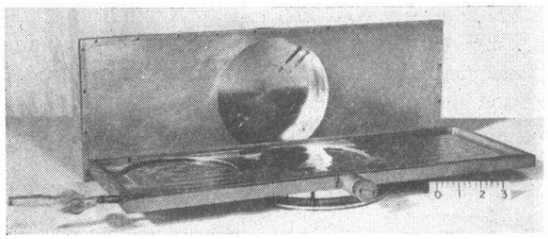

FIG. 1. View of the beta-counter disassembled.