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THE RADIOACTIVE DECAY OF POTASSIUM 40

by

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CHAPTER I

INTRODUCTION

Potassium 40 is one of the six known radioactive isotopes occurring in nature with an atomic weight below 206, that is, below the uranium-radium, uranium-actinium, and thorium families of radioactive elements.[1] These radioactive isotopes are listed in Table 1, with their natural isotopic abundance, type of radiation emitted, and approximate half-life.

Not only is K^{40} the radioactive isotope of lowest atomic weight occurring in nature, it is also one of the most abundant. In ordinary potassium, the isotopic abundance of K^{40}, as determined by Nier in 1950, is .0119 ± .0001 percent;[2] the abundance of potassium in the earth's crust is about 2.6 percent. The abundance of K^{40} in the earth's crust, therefore, is about 2×10^{-4} percent, and this is exceeded only by that of thorium and uranium: 12×10^{-4} percent and 4×10^{-4} percent, respectively.[3]

1. Early Investigations of K^{40}

The radioactivity of potassium was first observed in 1905 by J. J. Thomson,[4] and was definitely established in 1906 by Campbell and Wood,[5] who measured the activity of several different potassium salts with an ionization chamber. As samples, they used salts of potassium that had come from different parts of the earth and had been specially purified. The fact that the same activity was observed in all samples demonstrated

TABLE 1

NATURAL RADIOACTIVE ISOTOPES BELOW ATOMIC WEIGHT 206

Isotope	Isotopic abundance percent	Type of radiation emitted	Half-life years
$_{19}K^{40}$	.012	β, γ, K	1.3×10^{9}
$_{37}Rb^{87}$	27	β, γ	6×10^{10}
$_{60}Nd^{150}$	5.6	β	5×10^{10}
$_{62}Sm^{148}$	14	α	1.5×10^{11}
$_{71}Lu^{176}$	2.5	β, γ	7×10^{10}
$_{75}Re^{187}$	62	β	4×10^{12}

that the activity was characteristic of potassium and was not attributable to impurities. Campbell and Wood observed the radioactivity of rubidium, also, and noted the absence of radioactivity in the other alkali metals: lithium, sodium, and cesium.

Notable contributions to the knowledge of potassium radiation were made by Kolhörster,[6] who, in 1928, pointed out the existence of gamma-radiation, as well as beta-radiation, in potassium, and by Mühlhoff,[7] who, in 1930, showed that in the decay of potassium, beta-rays were much more abundant than gamma-rays. Mühlhoff also made the first careful measurements of the potassium half-life and gamma-ray energy.

In 1936, Yukawa[8] had proposed on theoretical grounds, that a nucleus might capture one of its own orbital K electrons, and it was pointed out by v. Weizsacker,[9] in 1937, that the existence of this type of K^{40} decay to A^{40} would account for the fact that the amount of argon found in the atmosphere is in excess of that to be expected on the basis of the abundances of the other inert gases.

That the radioactivity of potassium is due to the isotope of mass 40 was established in 1937 by Smythe and Hemmindinger,[10] who measured the activity of the three isotopes of potassium, which had been separated in a mass spectrograph. Only K^{40} was found to be radioactive.

The nuclear spin of K^{40} was measured in 1941 by Zacharias,[11] who used the molecular-beam method. The spin was found to be 4. Since the beta-decay product is Ca^{40}, which has an even number of both protons and neutrons, its spin is

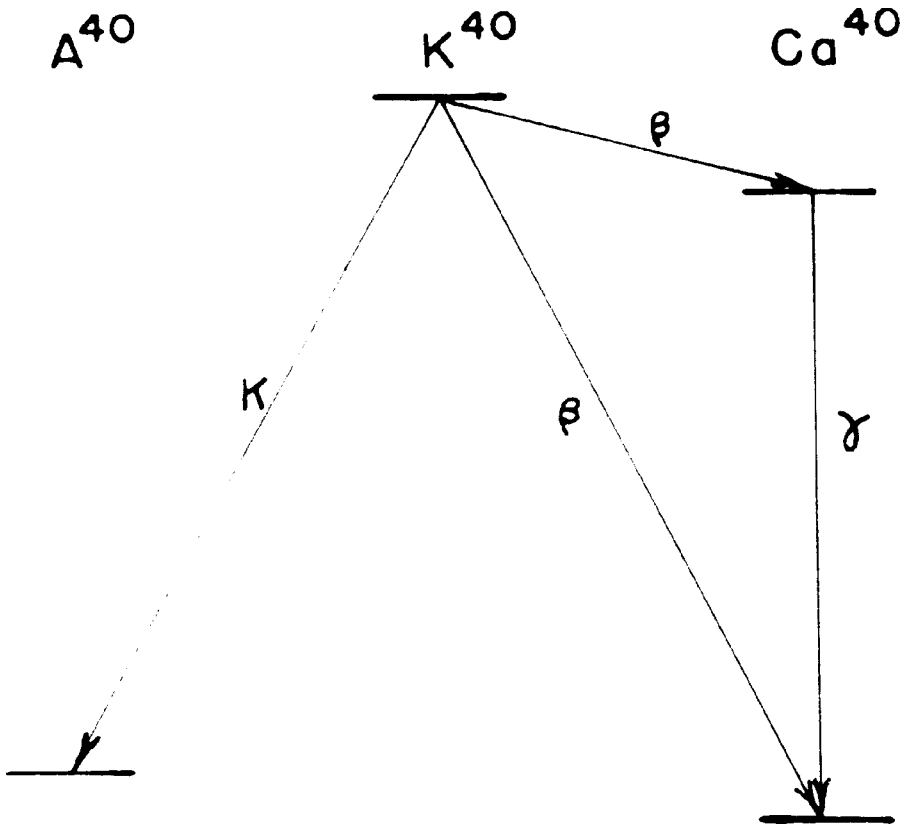
probably zero.[12] Therefore, the spin change in the transition is 4, for which the theory predicts a degree of forbiddenness of at least 3.[13] This high degree of forbiddenness indicates a very long lifetime, which, of course, is borne out by experiment.

2. Beta-and Gamma- Energies

Several measurements made before World War II[7,14-16] had indicated that the gamma-ray energy in K^{40} was greater than its beta-ray energy. In order to verify this fact, Hirzel and Wäffler,[17] in 1946, made a careful measurement of the beta-ray and gamma-ray energies. They measured the gamma-ray energy by the coincidence-counter method, taking an absorption curve of the secondary Compton electrons created by the gamma-rays, and they measured the beta-ray energy by taking an absorption curve and comparing it with the absorption curve of other beta-emitters of known energy. They concluded that the maximum beta-ray energy was 1.41 ± .02 Mev and that the gamma-ray energy was 1.54 ± 0.1 Mev. If the beta-ray energy had been found to be larger than the gamma-ray energy, it would have been possible to assume two beta-rays, one of low energy going to an excited state of calcium, and the other going directly to the ground state. The gamma-ray would then correspond to the transition from the excited state to the ground state of calcium, in cascade with the low-energy beta-ray (Fig. 1). It would be necessary to assume the two beta-rays because Mühlhoff had shown that there were many more beta-ray than gamma-ray disintegrations. However, since the gamma-ray energy was found to be greater, this possibility was

Figure 1

Early Decay Scheme Proposed for K^{40}



ruled out, and Hirzel and Wäffler proposed that the gamma-rays be placed in cascade with the electron-capture decay (Fig. 18), although there was still no direct evidence for the existence of electron capture in K^{40}.

With two exceptions, subsequent measurements of the beta-ray and gamma-ray energies have confirmed the earlier conclusions that the gamma-ray energy is greater, although the energy difference is small. The results of these measurements are given in Tables 2 and 3. It should be noted that the two measurements of the beta-ray energy which are unusually high and which would place the beta-ray energy higher than the gamma-ray energy are probably in error. One of these, the work of Franchetti and Giovanozzi,[20] was made with a cloud chamber, where multiple scattering of the beta-rays can distort the cloud-chamber tracks enough to simulate the very few high-energy tracks that are necessary to raise the apparent maximum energy. The other, the 1948 measurement of Floyd and Borst,[21] giving 1.9 Mev, was declared probably inaccurate by Floyd and Borst themselves, who published a later paper in which they reported a new measurement.

In the light of these measurements, which are in relatively good agreement, the most probable values of the beta-ray and gamma-ray energies can be given as $E_{\beta\ max.} = 1.38 \pm .05$ and $E_{\gamma} = 1.50 \pm .05$. It may be noted that if the two doubtful measurements are disregarded, every gamma-energy value is above every beta-energy value and that, with the exception of the later value of Floyd and Borst, there is no overlap within the probable errors. In the work described in this thesis

TABLE 2

BETA-ENERGY DETERMINATIONS

Observer	Year	Method	Maximum Energy (Mev)
Hirzel & Wäffler[17]	1946	Absorption	1.41 ± .02
Dželepow et. al.[18]	1946	Spectrograph	1.35 ± .05
Henderson[19]	1947	Absorption	1.3
Franchetti & Giovanozzi[20]	1948	Cloud Chamber	1.7 ± .1
Borst & Floyd[21]	1948	Absorption°	1.9 ± .2
Alburger[22]	1949	Spectrograph°	1.40 ± .03
Floyd & Borst[23]	1949	Absorption°	1.45 ± .15
Bell, Weaver & Cassidy[24]	1950	Scintillation Spectrograph°	1.36 ± .05

° Enriched source from Carbide and Carbon Chemical Corp., Oak Ridge, Tennessee.

TABLE 3

GAMMA-ENERGY DETERMINATIONS

Observer	Year	Method	Energy (Mev)
Mühlhoff[7]	1930	Absorption	1.47
Behounek[14]	1931	Absorption	1.52
Hirzel & Wäffler[17]	1946	Absorption	1.54 ± .1
Gleditsch & Graf[25]	1947	Absorption	1.55 ± .05
Bell & Cassidy[26]	1950	Scintillation Spectrograph	1.46 ± .04
Pringle, et. al.[27]	1950	Scintillation Spectrograph	1.47 ± .03

no attempt was made to repeat the energy determinations.

3. Half-Life Determinations

The question of the half-life of K^{40} and the relative number of beta-, gamma-, and electron-capture-disintegrations occurring in its decay has by no means been settled. The results of seven recent determinations of the half-life for beta-decay are listed in Table 4. All of these measurements were made with Geiger counters in a calibrated geometry, and all except that of Stout[31] are subject to the criticism that large corrections had to be applied for the solid angle subtended by the counter and for the absorption of the radiation in the counter windows. In the measurements, for example, of Hirzel and Wäffler[30] and of Floyd and Borst,[21,23] the solid angle was about 20 percent and, in most cases, correction for window absorption was as much as 10-20 percent. In the measurement of Bleuler and Gabriel,[28] mounting the source on the inside wall of the counter eliminated window absorption, but making the heavy brass wall of the counter thus serve as the source backing undoubtedly caused a great deal of back-scattering of the radiation. The apparent failure of Bleuler and Gabriel to make correction for this effect probably accounts for the high specific activity they report. The most ingenious, and probably the most reliable, method used was that of Stout 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

TABLE 4

BETA HALF-LIFE DETERMINATIONS

Observer	Year	Half-Life (Years)
Mühlhoff[7]	1930	18×10^8
Bramley & Brewer[15]	1938	$15.2 \pm 3.0 \times 10^8$
Bleuler & Gabriel[28]	1947	$7 \pm 1 \times 10^8$
Graf[29]	1948	$14.2 \pm 0.6 \times 10^8$
Borst & Floyd[21]°	1948	$17.8 \pm 1.8 \times 10^8$
Hirzel & Wäffler[30]	1948	$12.0 \pm 1.9 \times 10^8$
Stout[31]	1949	$13.1 \pm 1.5 \times 10^8$
Floyd & Borst[23]°	1949	$16.1 \pm 1.9 \times 10^8$
Spiers[51]	1950	13.4×10^8

° Enriched source from Carbide and Carbon Chemical Corp., Oak Ridge, Tennessee.

corrections.

Measurements of the gamma-ray activity have also shown considerable variations, as is shown in Table 5. In all cases, the activity of massive samples of potassium salts was measured with counters (an ionization chamber in the case of Behounek[14]) which had been calibrated with other radioactive elements. The unusually low value of Mühlhoff[7] was apparently due to his failure to consider absorption of the radiation in the source itself.

In 1943, Thompson and Rowlands[33] during cloud-chamber measurements on K^{40}, made the first observation of electron-capture decay. They found many very short tracks which they attributed to low-energy photoelectrons ejected from the counter gas by the x-rays emitted following the electron-capture decay, as the vacant K shell of the resulting argon atom was filled. They estimated the ratio of electron capture to beta-ray emission to be 3 or 4 to 1. In 1947, using a counter arranged to admit gaseous absorbers, Bleuler and Gabriel[28] undertook to measure the intensity of the x-rays resulting from the electron capture in K^{40}. As a result of this experiment, the K/beta ratio was estimated to be 1.9/1. Several measurements have been made of the amount of argon trapped in potassium-bearing minerals, and on the assumption that all the argon had been produced by decaying potassium, the K/beta ratio was computed. By this method, Ahrens and Evans[32] obtained 1.38/1, Floyd and Borst[21] about 1/1, and Suess,[34] .05/1. Aldrich and Nier,[35] in a refinement of this method, used the mass spectrograph to analyze the ratio of

TABLE 5

GAMMA-ACTIVITY DETERMINATIONS

Observer	Year	Gamma-rays/gm K/sec.
Mühlhoff[7]	1930	.97
Behounek[14]	1931	3.8
Gleditsch & Graf[25]	1947	3.6 ± .8
Ahrens & Evans[32]	1948	3.3 ± .1
Hirzel & Wäffler[30]	1948	2.4 ± .3
Floyd & Borst[23]	1949	1.4 ± .3
Spiers[51]	1950	3.0

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 the K/beta ratio was estimated to be about 0.10/1. It can be seen that the discrepancies here are enormous, the estimates varying from 5 to 190 electron-captures per 100 betas. These discrepancies can be attributed to the uncertainties inherent in the methods used in making the measurements. These experiments will be discussed more fully in Chapter IV and their uncertainties pointed out.

4. Geological Significance.

The widely differing values of the half-life that have been reported in the past few years have an interesting geological significance. Consider two possible sets of decay constants: (1) using the beta-activity and K/beta ratio proposed by Bleuler and Gabriel and about 3.6 gamma-rays/g K/sec; (2) using the beta-activity measured by Stout and the K/beta ratio of Aldrich and Nier, and again about 3.6 gamma-rays/g K/sec. Now consider the heating in the earth's crust caused by K^{40} as given by these two decay schemes. The heating is due only to the beta- and gamma-rays; the energy of the K-capture decay is carried off by a neutrino which is presumably not absorbed in the earth,[36] and the heat contributed by the energy of the atomic x-ray following K-capture is so small that it may be neglected here. But while the K-capture does not contribute to the heating, the number of disintegrations going by this path does influence the total half-life, which, in turn, determines the number of K^{40} atoms that were available for decay in ancient geological time.

The heating of the earth's crust due to K^{40} under the two schemes, 1 and 2, is tabulated in Table 6. The first four lines give the present beta- and gamma- activities and corresponding heating on the basis of the two schemes; line 5 gives the total present-day heat production; lines 6 and 7 the proposed electron capture to beta-activity ratio and corresponding total half-life. Then for every half-life one goes back in the earth's history, the heating is doubled. Lines 8 and 9 give the total heating for the two schemes 3.3×10^9 and 2.0×10^9 years ago, which are, according to the best age determinations by radioactive methods,[37,38] the time of the formation of the earth, and the time at which the earth had a solid crust, respectively.

Although the heating under scheme 1 is at present only 1.5 times that under scheme 2, 3.3×10^9 years ago the heating under scheme 1 would have been 4000 times that under scheme 2. According to estimates of geologists,[32,39] this heat would have been sufficient to prevent the earth's having a solid crust 1.5×10^9 years ago, whereas present-day estimates put the time of the earth's becoming solid at about 2×10^9 years ago. Furthermore, geologists estimate that if the number of decays by electron capture in K^{40} were as large as proposed by Bleuler & Gabriel, the abundance of potassium-derived argon in the earth's atmosphere would be much greater than the observed abundance of all argon found in the atmosphere. The findings of the geologists, then, contribute to the evidence that the activity reported for K^{40} by Bleuler and Gabriel is too large.

TABLE 6

HEATING OF EARTH BY K^{40}

	Bleuler & Gabriel	Stout, Aldrich & Nier
Beta-rays/g K/sec. of Avg. Energy = .49 Mev.	52.5	30
Heat released/g K by beta rays = .49 x line 1	26 Mev/sec	14.7 Mev/sec
Gamma-rays/g/sec. of energy = 1.5 Mev	3.6	3.6
Heat released/g by gamma-rays = 1.5 x line 3	5.4 Mev/sec	5.4 Mev/sec
Total heat production/g K by beta-and gamma-rays	31.4 Mev/sec or 38 Cal/yr.	20.1 Mev or 24 Cal/yr.
K/B ratio	1.9	0.1
Total half-life	$.24 \times 10^9$ yr.	1.2×10^9 yr.
Heating 2.0×10^9 yrs ago. Cal/g K/year	12,500	77
Heating 3.3×10^9 yrs ago. Cal/g K/year	610,000	160

5. Problem Posed

In view of the large discrepancies between the values that have been reported for the decay constants of K^{40}, it seemed worth while to attempt more accurate determinations of its half-life and the relative numbers of beta-rays, gamma-rays, and electron captures occurring in its decay. To make these measurements, it was decided to develop special counters adapted to the measurement of weak activities. The measurements with these counters will be described in the following chapters.

CHAPTER II

BETA-RAY MEASUREMENTS

It is, or course, impossible to determine the half-life of potassium^{40} by following the decay of its activity, since the half-life is of the order of 10^9 years. Instead, it is necessary to measure its decay constant, λ : the number of disintegrations per atom per second. The decay constant is defined:

$$dN = -N \lambda \, dt \qquad (1)$$

where N is the number of atoms available for disintegration, and dN is the number decaying in time, dt. Integrating this equation gives the usual expression for the number of atoms remaining out of an original number, No, after a time, t, has elapsed:

$$N = No \, e^{-\lambda t} \qquad (2)$$

The half-life is defined as the time required for N to fall to the value $\frac{No}{2}$, and if this value is put into equation 2, the relation between the decay constant and the half-life is obtained:

$$T_{\frac{1}{2}} = \frac{.693}{\lambda} \qquad (3)$$

1. Difficulties in Beta-Activity Measurement

Measuring the activity of K^{40} is difficult because the activity of the available sources is weak. K^{40} has not been obtained in pure form because it is hard to separate from the other isotopes of potassium. Until recently, it has been

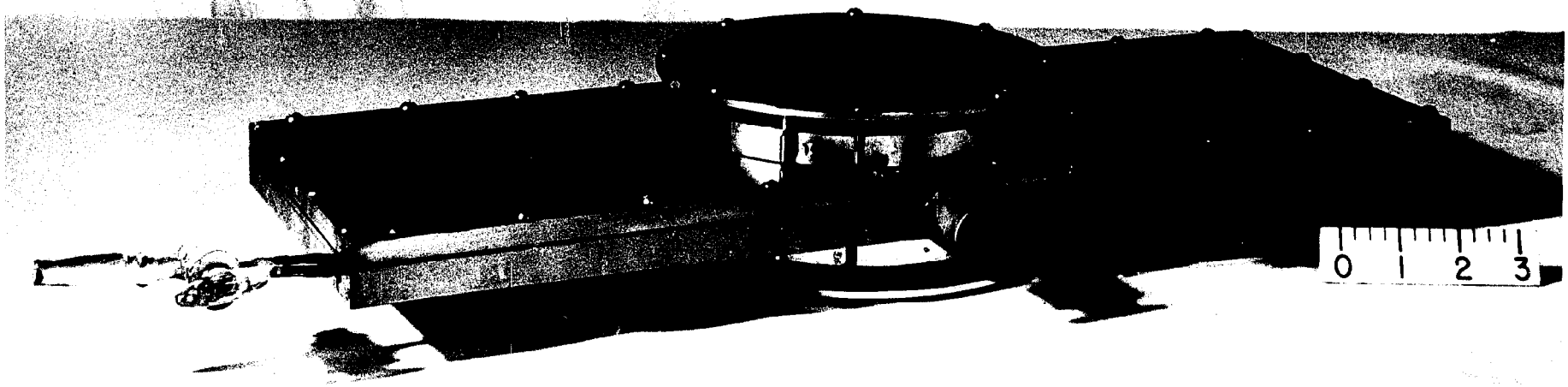
necessary to use, as sources, salts of ordinary potassium containing only about one part of K^{40} in 10,000 parts of potassium. In the past two years, limited amounts of potassium salts enriched in K^{40} have been made available by the Atomic Energy Commission through the Carbide and Carbon Chemicals Corporation, Oak Ridge, Tennessee. These samples contain isotopic abundances of K^{40} as high as 1.3 percent.

To attain measurable activities with the weak sources available, large amounts of the material must be used, and since to prevent appreciable absorption of the beta-rays, the sources must be kept thin -- a few mg/cm^2 --, the material must be spread over a large surface area. Conventional cylindrical Geiger counters, with the source outside the counter, are not well suited to measurements on sources of large area, because a thin window has to be provided to admit the beta-rays and it is not possible to use windows that are both thin and large, since they must be capable of withstanding atmospheric pressure. At best, some of the beta-rays will be absorbed by any window, and this absorption interferes with accuracy when absolute numbers of disintegrations are to be determined.

Obviously, the best possible arragnement 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, using an adaptation of the cell counters developed by Beyster and Wiedenbeck[40]. This counter is shown in Fig. 2.

Figure 2

Sliding-Tray Beta-Counter

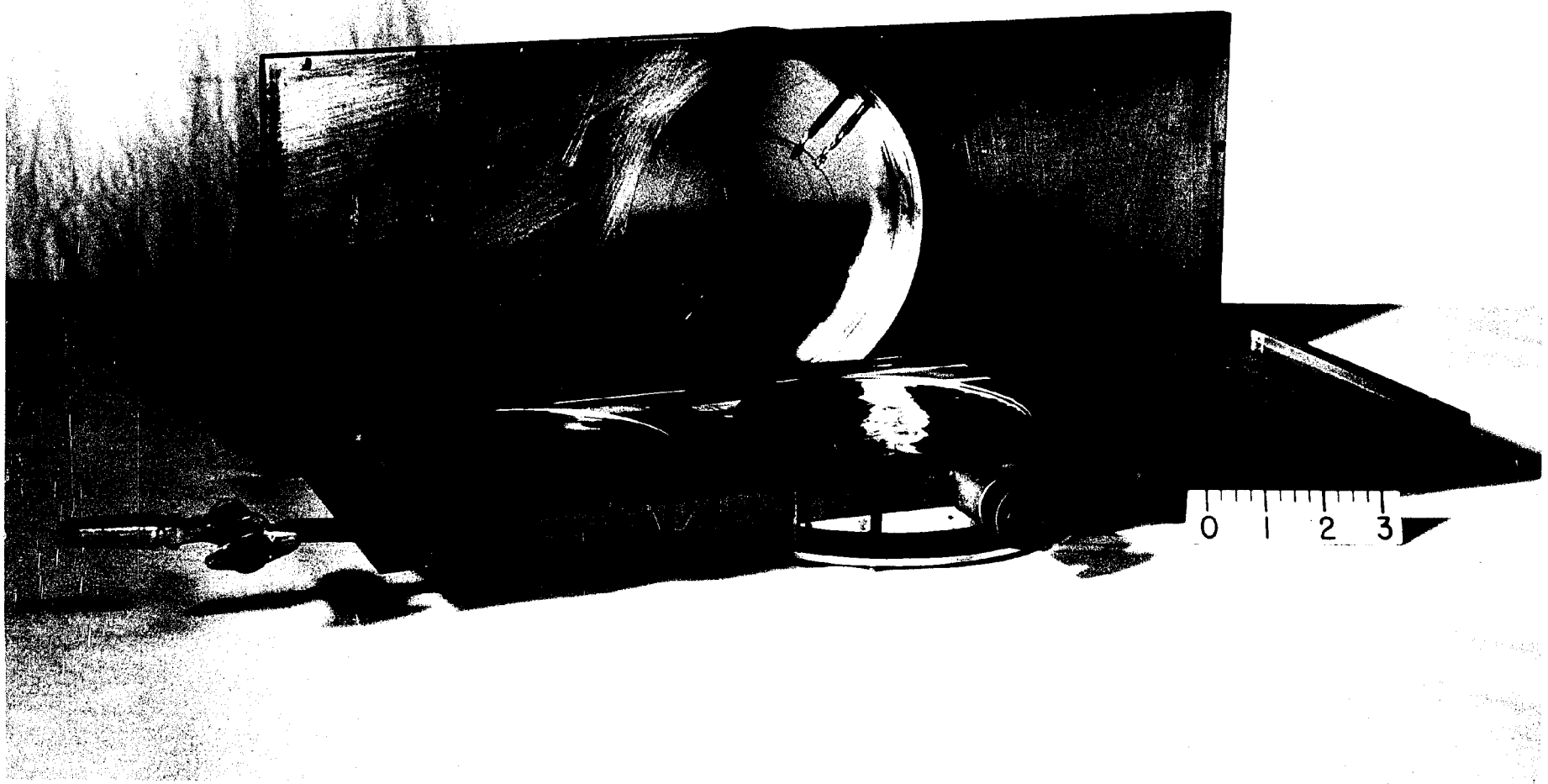

0 1 2 3

2. Description of the Beta-Counter

The counter uses two independent, pancake-like cell sections. Basically, each section is a short, brass cylinder, 6 in. in diameter and 1 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 electrical connection to the anode wire is made by a tungsten wire sealed into one of the glass supporting rods. The two cells are then placed together, with a common cathode surface between them, and operated as a single counter. This common cathode surface is a thin aluminum foil disc on which the source of K^{40} is mounted. The counter is arranged so that, for background measurement, the source-bearing foil may be replaced by a blank foil without breaking the vacuum seal of the counter. The foil-changing mechanism and construction of the counter can be seen in Fig. 3. The two aluminum foils, one blank, the other bearing the source, are mounted on a brass tray 3/16 in. thick. The tray may be moved back and forth by a rack-and-pinion-arrangement, so that either foil may be centered in the counter. The other foil moves out into one of the wings of the counter; each wing has a recess just large enough to accommodate the sliding tray. The pinion gear, which imparts the motion to the sliding tray, is turned by a knob from outside the counter. The shaft carrying the knob and pinion gear is the inner part of a tapered, brass, ground joint resembling a stopcock. This ground joint is lubricated with

Figure 3

Beta-Counter Dissassembled


0 1 2 3

stopcock grease and provides the vacuum seal for the rotary motion. The joints between all parts of the counter are made vacuum tight with rubber gaskets -- strips of 1/16 in. square cross-section, partially recessed in grooves along the surfaces to be sealed, as shown in the figure. Very little trouble was experienced with leaks through these gaskets or through the rotating ground joint.

3. Advantages of the Beta-Counter

The geometrical arrangement of the beta-counter described here affords the following advantages over conventional counters:

1. Sources of considerable area can be used (about 120 cm^2), and every beta-ray which leaves the source enters the sensitive volume of the counter. Particles which leave the front surface of the source pass into the top cell of the counter, and particles which are directed down into the aluminum backing pass into the bottom cell of the counter through the aluminum foil. Absorption of the particles in the aluminum foil and in the source material itself, however, must be taken into account, as usual.

2. No correction for back-scattering is necessary. It makes no difference whether a particle directed into the backing passes through into the bottom section or is scattered back into the top section, since it will be counted in either case unless it is completely absorbed in the source.

4. 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 the plateau, sensitivity, and efficiency of the counter.

Plateau and Operating Conditions. The counter was operated with a scale-of-256 circuit and mechanical register. The scaler used was a commercial unit made by the Instrument Development Laboratories (model 161). A gas mixture of 90 percent argon and 10 percent alcohol was used in the counter and the counter was filled to 10 cm pressure with this mixture. Although the counter was self-quenching, an auxiliary quench circuit (Instrument Development Laboratories, model 101-1) was always used because, with it, the plateau was flattened considerably. The plateau of the counter was about 125 volts long, with a rise in counting rate along the plateau of about 3 percent per 100 volts. Fig. 4 shows a typical plateau curve for the counter, taken with the two sections operated in parallel -- the usual operating condition. The plateau of the counter was checked frequently throughout the course of the experiments. The counter was operated at about 60 volts above threshold, approximately in the center of the plateau.

Sensitivity. Tests were made to determine whether the counter was uniformly sensitive over its entire area. An aluminum blank holding a point-source of Tl^{204}, a pure beta-emitter, was placed on the sliding tray inside the counter so that it was possible to move the point-source across the

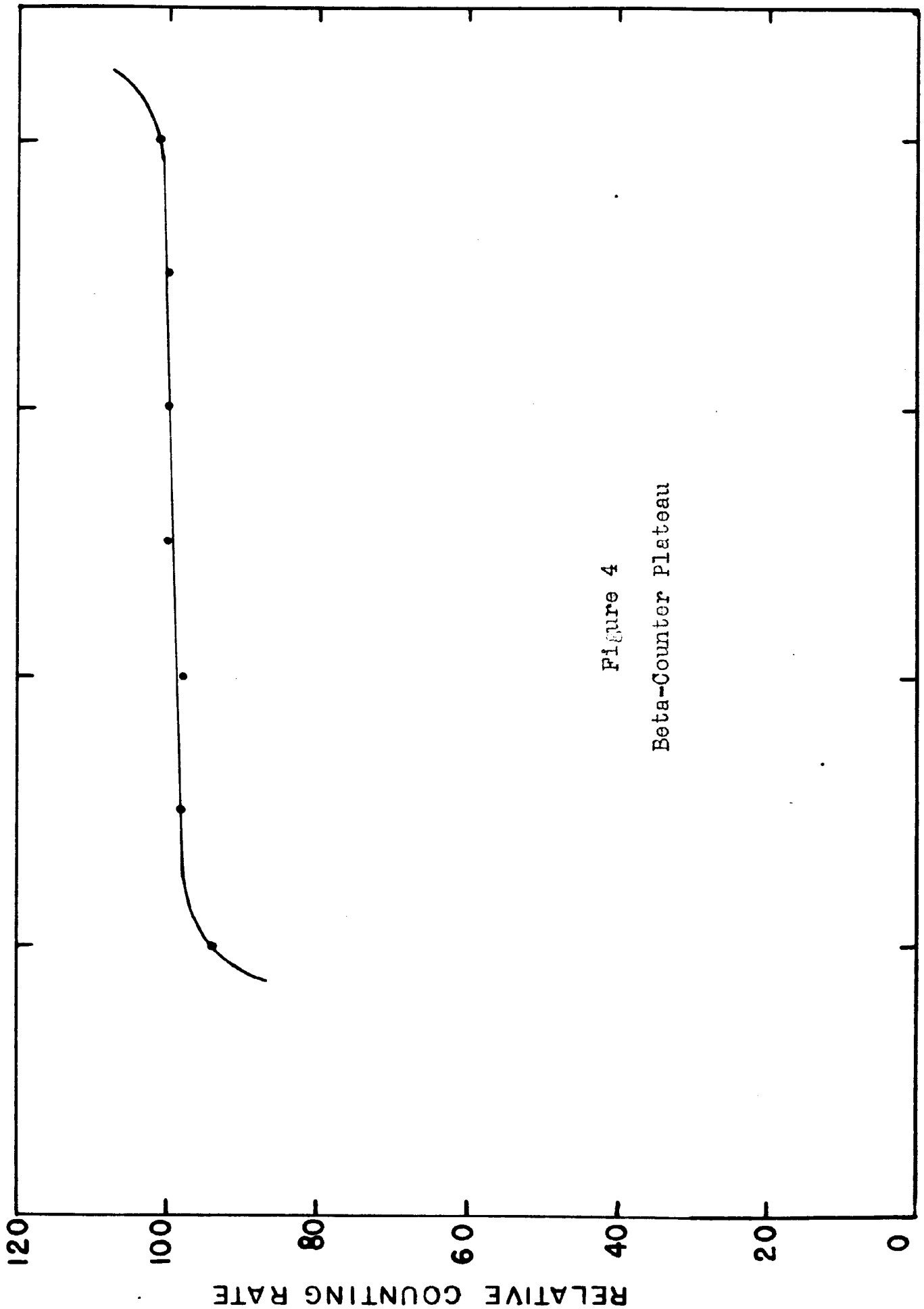


Figure 4

Beta-Counter Plateau

counter. The counting rate was measured as the source was moved in 1/4 in. steps across the diameter, from one wall of the cylindrical counter cell to the other. The results are shown in Fig. 5. The small dip in counting rate at the center of the counter is not surprising, because a particle leaving the source at the center and passing vertically up through the counter cell passes through a region of zero field.

Two further tests were made with collimated sources. First, a small brass shield, heavy enough to stop the beta-rays, was placed over the Tl^{204} source in the counter so that the beta-rays were restricted to entering the counter laterally along the tray, in a half cone about 30 degrees high. Second, a thin, mica window was placed in the cover-plate of the counter so that a vertically collimated beam of beta-rays could be fired down into the counter in a cone about 10 degrees wide. The results of these two tests are shown in Fig. 6. The fact that the central dip is more pronounced in the vertically collimated case than in the uncollimated case seems conclusive evidence that particles passing vertically through the center of the counter in the region of zero field are not counted . The effect of this lack of sensitivity on the total counting rate of an extended source is negligible, however, since only particles leaving the exact center of the source in a vertical direction are missed, and, in fact, the curve for the uncollimated point source (Fig. 5) shows that those missed actually amount to only about 5 percent of the particles from the central point.

It should be noted that the diameter of the sources used

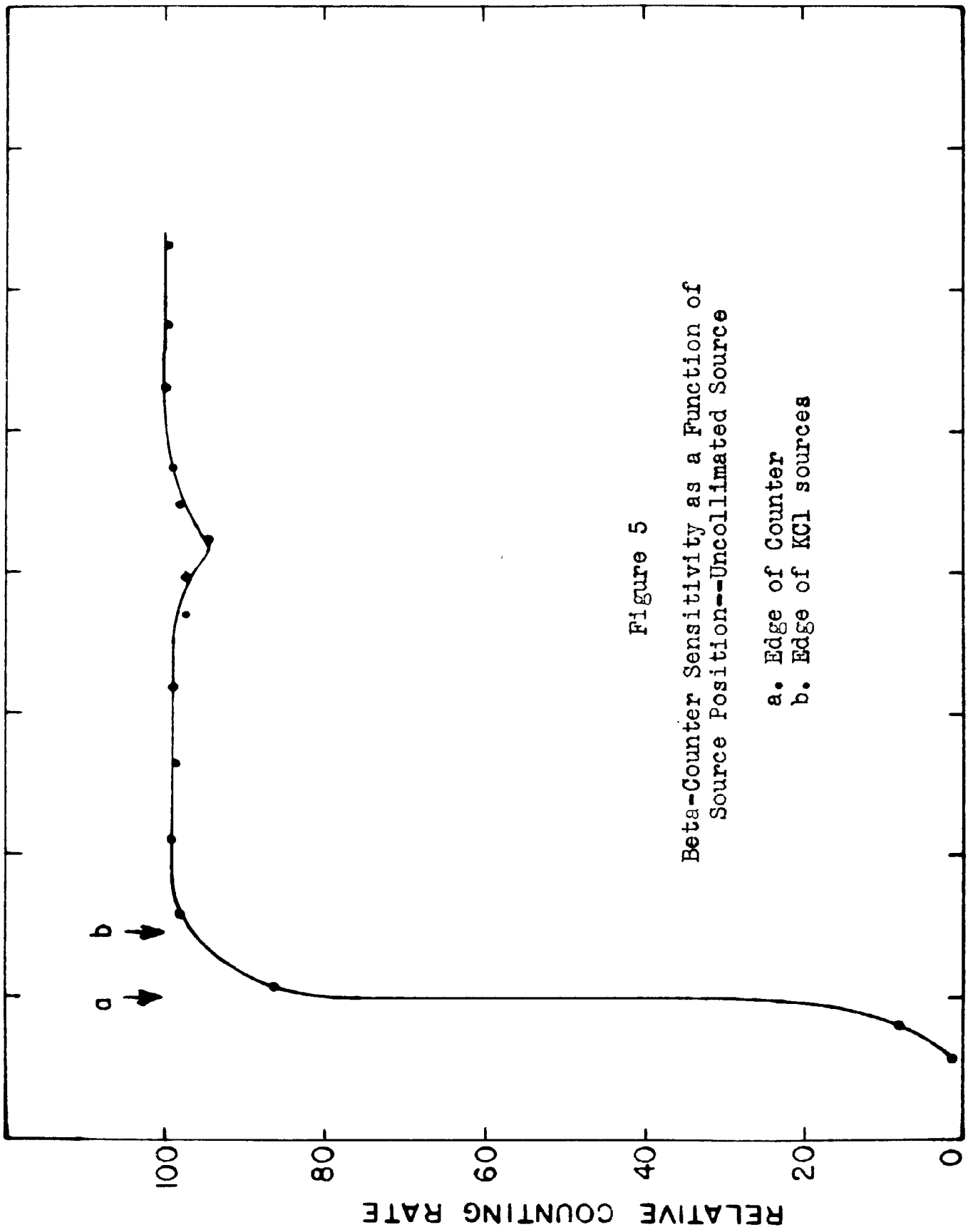


Figure 5

Beta-Counter Sensitivity as a Function of Source Position--Uncollimated Source

a. Edge of Counter
b. Edge of KCl sources

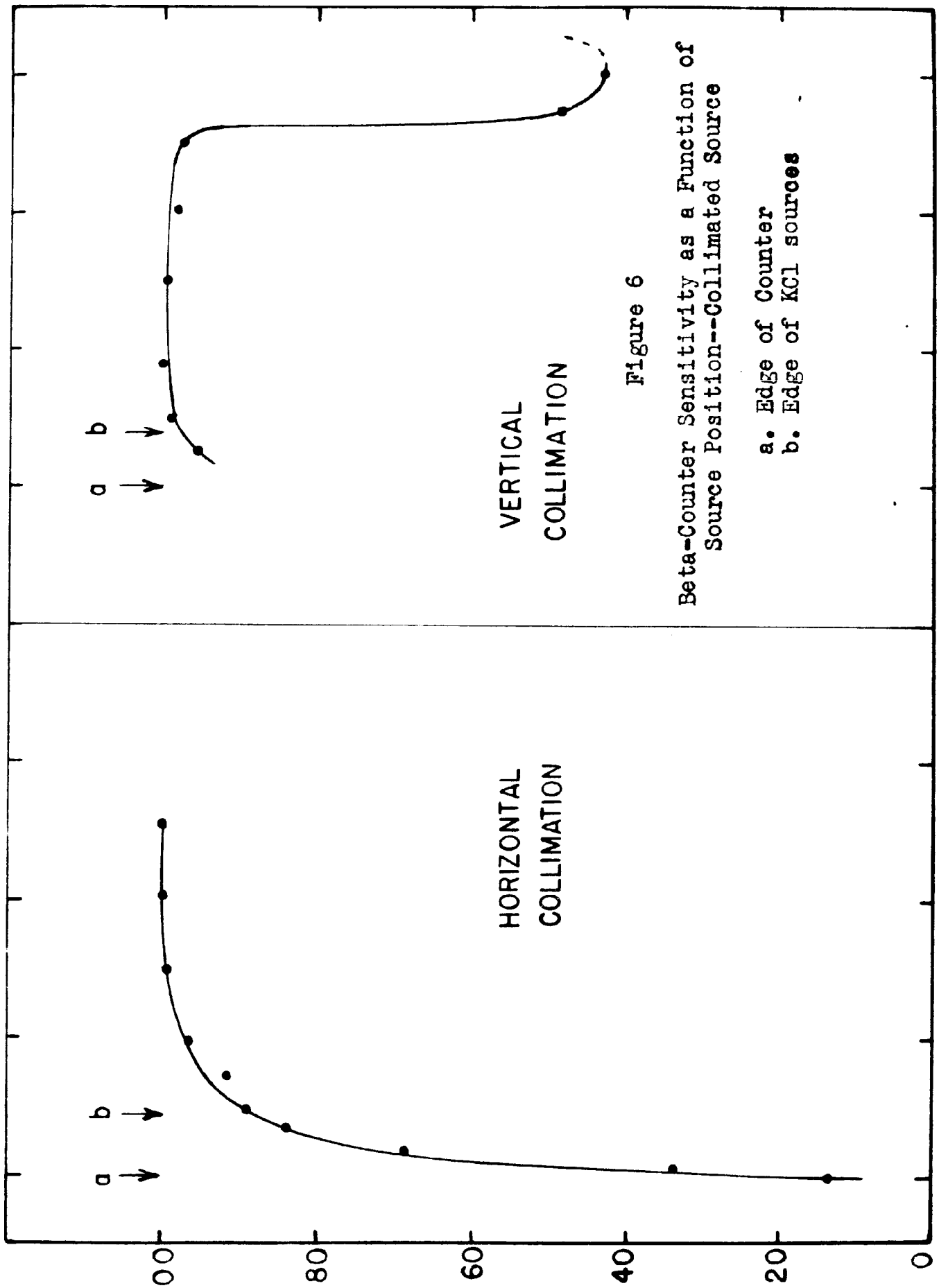


Figure 6

Beta-Counter Sensitivity as a Function of Source Position--Collimated Source

a. Edge of Counter
b. Edge of KCl sources

in the measurements on K^{40} was smaller than the inside diameter of the counter cells, so that about a 3/8 in. space was left between the edge of the extended source and the wall of the counter. The position of the edge of the K^{40} sources is indicated in Fig. 5 and Fig. 6; it can be seen that enough space was allowed to keep the sources out of the region of rapidly falling sensitivity at the edge of the counter.

The extended sources of K^{40} used in the counter did extend slightly into the region of lowered sensitivity at the edge of the counter and a correction to the counting rate of about 2 percent was necessary to allow for this lack of sensitivity, and to allow for the fact that beta-rays directed laterally within the source would not escape into the counter to be detected.

Efficiency. A measurement of the efficiency of the counter was made by the coincidence method. A small, thin-walled, glass counter was placed over a mica window in the top plate of the sliding-tray counter so that beta-rays from a point source of Tl^{204} could pass down through the thin-walled counter and the two sections of the big counter. The two sections of the sliding-tray counter and the thin-walled counter above it were operated independently, each with its own scaler, and coincidences between the counters were recorded with a Rossi-type coincidence circuit.

A determination of the efficiency of the center counter of the group of three in this telescope arrangement requires only a measurement with the same source, of the triple coincidences between all three counters and the double coincidences

between the two outer counters, without regard to the fraction of the beta-rays that enter each counter. The number of triple coincidences, N_{123}, will be: the number of disintegrations of the source, times the fraction, F, that passes through all three counters, times the fraction of these that are counted in each counter, or, in other words, the efficiency, ϵ, of each counter. Thus:

$$N_{123} = N F \epsilon_1 \epsilon_2 \epsilon_3 \qquad (4)$$

Similarly, the number of double coincidences, N_{13}, is:

$$N_{13} = N F \epsilon_1 \epsilon_3 \qquad (5)$$

where F, the fraction passing through the two counters, is the same as in the triple coincidence case, since the geometry is unchanged and the fraction passing through the three counters is the same as the fraction passing through the two outside ounters. The efficiency of the center counter is then obtained by dividing the triple coincidences by the double coincidences:

$$\frac{N_{123}}{N_{12}} = \frac{N F \epsilon_1 \epsilon_2 \epsilon_3}{N F \epsilon_1 \epsilon_3} = \epsilon_2 \qquad (6)$$

In practice, it is better to measure, not the triple coincidences, but the anticoincidences, that is, the events in which counters 1 and 3 fire but counter 2 fails to fire. The number of anticoincidences, N_a, depends on the fraction of the particles which counter 2 fails to count, or $1 - \epsilon_2$, and so the number of anticoincidences recorded is:

$$N_a = N F \epsilon_1 \epsilon_3 (1 - \epsilon_2) \qquad (7)$$

The efficiency in this case is one minus the ratio of coin-

cidences:

$$1 - \frac{N_a}{N_{13}} = \epsilon_2 \qquad (8)$$

The advantage in measuring anticoincidences instead of triple coincidences lies in the fact that if the efficiency to be measured is near 100 percent, better statistical accuracy can be obtained with the same expenditure of time.

When the beam of beta-rays passing through all three counters was collimated so that they passed close to the anode wires of the sliding-tray counter, the efficiency of each section of the sliding-tray counter was found to be 98 percent. When the beam of beta-rays passed far from the wire of the sliding-tray counter, it was found that the measured efficiencies were much less -- as low as 30 percent. However, this drop, which was expected, did not mean that the counter efficiency was low at these points. Bradley and Wiedenbeck[41] have demonstrated that in this type of cell counter, when the particle passes far from the anode wire, there is often a delay of several micro-seconds between its passage and the formation of the pulse on the wire. Now, since the resolving time of the coincidence circuit used was 0.8 microseconds, any pulse delayed by more than this interval in counter 2 will cause an anticoincidence to be recorded, as though counter 2 had failed to fire, and hence will lead to a false indication of lack of efficiency. The measured efficiency of 98 percent near the wire, together with the nearly uniform sensitivity across the counter, indicates that the overall efficiency of the counter is at least 98 percent, except in the unimportant

regions of lowered sensitivity described above. That the long delays in these counters have no effect on their efficiency is confirmed by the measurements on cell counters made by Bradley and Wiedenbeck.[41] Since occasional long delays are observed even when the particles pass near the anode wire, the measured figure of 98 percent at the wire is a minimum value.

It should be noted that in this efficiency measurement, the existence of spurious counts is not ruled out, for they would not be detected by the coincidence method. However, the flatness of the plateau for the counter is convincing evidence that no significant number of spurious counts occurs, since their number always increases as the counter voltage is increased, causing a steeply rising plateau.[42]

The efficiency thus demonstrated experimentally is confirmed by calculation from the known specific ionization of gases by beta-rays. If one ion pair is formed by a beta-ray in passing through the counter, a count will be produced. If x is the average number of ion pairs formed by a beta-ray in traversing the counter, then e^{-x} is the probability that no ion pairs at all are produced, and $1 - e^{-x}$ is the probability that at least one pair will be produced, and this expression represents the efficiency. The average number of ion pairs produced is the product of the specific ionization of the beta-rays in the counter gas, their path length in the counter, and the gas pressure in atmospheres. For argon with high-energy electrons, the specific ionization is about 30 ions/cm/atmosphere.[43] With the gas pressure of 10 cm used in the sliding-tray counter, it may easily be calculated that the efficiency for a high-energy particle travelling completely through one

cell of the counter is 99.9 percent; for a particle travelling 1 cm in the counter, the efficiency is 98.6 percent. Since no beta-ray has a path length smaller than 1 cm in the counter, and most are much longer, and since low-energy particles produce a higher specific ionization, the average efficiency is at least 99.0 percent. A correction of one percent was made in the K^{40} measurements to allow for counter inefficiency.

Background. To reduce the background counting rate to as low a value as possible, the sliding-tray counter was surrounded by 3 in. of lead to shield it from external radiation. This shielding reduced the background counting rate for the two sections from about 13 counts/sec to 3.6/sec.

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

5. Measurements with Ordinary Potassium.

The actual measurements of the beta-ray activity of K^{40} were divided into two parts: measurements with ordinary potassium, and measurements with an enriched source.

Sources. For the measurements on ordinary potassium, sources of chemically pure potassium chloride were used. A saturated solution of KCl in water was made up, and a special atomizer was used to spray it onto aluminum-foil blanks in a fine mist; when dry, the KCl was found to adhere very well to the aluminum surface. Any desired thickness could be deposited, and the amount deposited was determined by the accurate weighing of the blank both before and after spraying. There was a

tendency for the deposit to crystalize in tiny lumps on the surface of the aluminum, and it was estimated that there may have been variations of a factor of two in the thickness of a source, but, in the large amounts required, no practical way was found to deposit the KCl more evenly. The differences in measured activity caused by the varying absorption of different parts of the source were insignificant, however, and caused no serious error.

Method of Measurement. Twenty sources were made of thicknesses varying from .5 to 13 mg/cm^2 and on three weights of aluminum-foil backing: 5.43, 2.67, and 0.18 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 measurements every 1000 sec. In practice, it was found that the sources needed to be covered with an aluminum foil. If this was not done when the thick sources were used, 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 which disrupted the normal counting action in some unknown manner. Accordingly, all sources were covered with aluminum foil, of the same weight as the backing foil.

Results. From the data obtained with various source- and backing-thicknesses, it was possible to extrapolate the measured activity to zero source-thickness and zero backing-thickness. The activity-measurement data are plotted in

Fig. 7, which shows specific activity against source-thickness for the three weights of backing. Fig. 8 shows the specific activity at zero source-thickness against backing-thickness. The extrapolated value obtained for the activity of KCl at zero source- and backing-thicknesses is 14.4 disintegrations/g KCl/sec. or 27.5 disintegrations/g K/sec.

No correction need be applied to this figure for the number of gamma-rays and electron captures that may have been counted. In the following chapters it will be shown that the gamma-ray and electron-capture intensities are each approximately 12 percent of the beta-activity, and since the efficiency of the sliding-tray counter for gamma-rays is less than 1 percent, only a negligible number of gamma-rays were counted. In the case of the electron captures, as will be explained in Chapter IV, the particles to be counted were 3 kev electrons, which have such a short range that they were unable to escape through the aluminum foils covering the source and were therefore not counted.

A three percent correction must be made to this value to allow for counter inefficiency and lack of sensitivity, as explained in the section on counter performance. The beta-activity of ordinary potassium is, therefore, 28.33 beta-rays/gm K/sec. This corresponds to a half-life for beta-decay in K^{40} of 14.2×10^{8} years, using .0119 percent[2] as the isotopic abundance of K^{40} in ordinary potassium.

Errors in Measurement. The probable error in the value for the abundance of K^{40} in ordinary potassium is ± 0.84 percent[2]. The statistical errors in the experiment itself include the

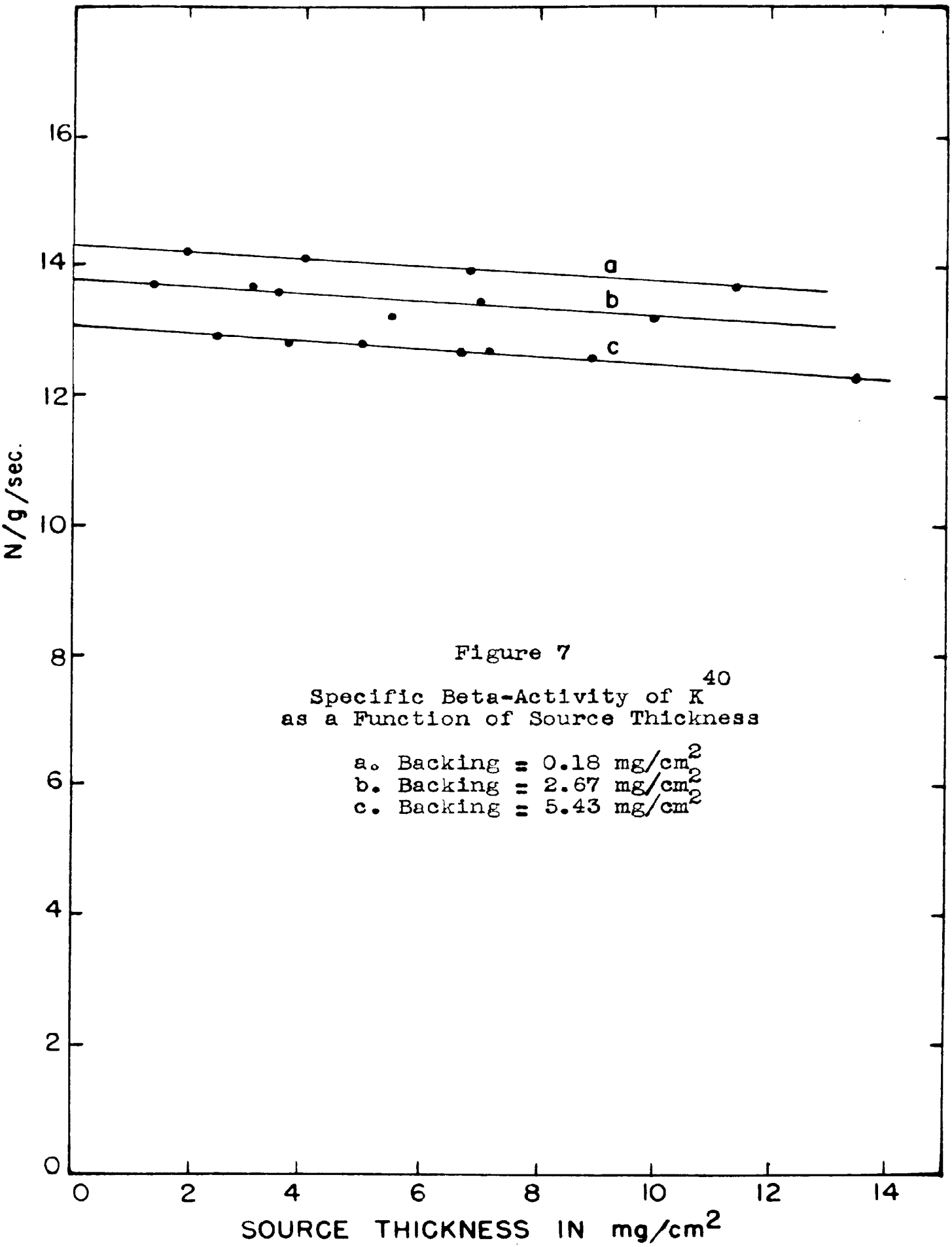


Figure 7

Specific Beta-Activity of K^{40}
as a Function of Source Thickness

a. Backing = 0.18 mg/cm^2
b. Backing = 2.67 mg/cm^2
c. Backing = 5.43 mg/cm^2

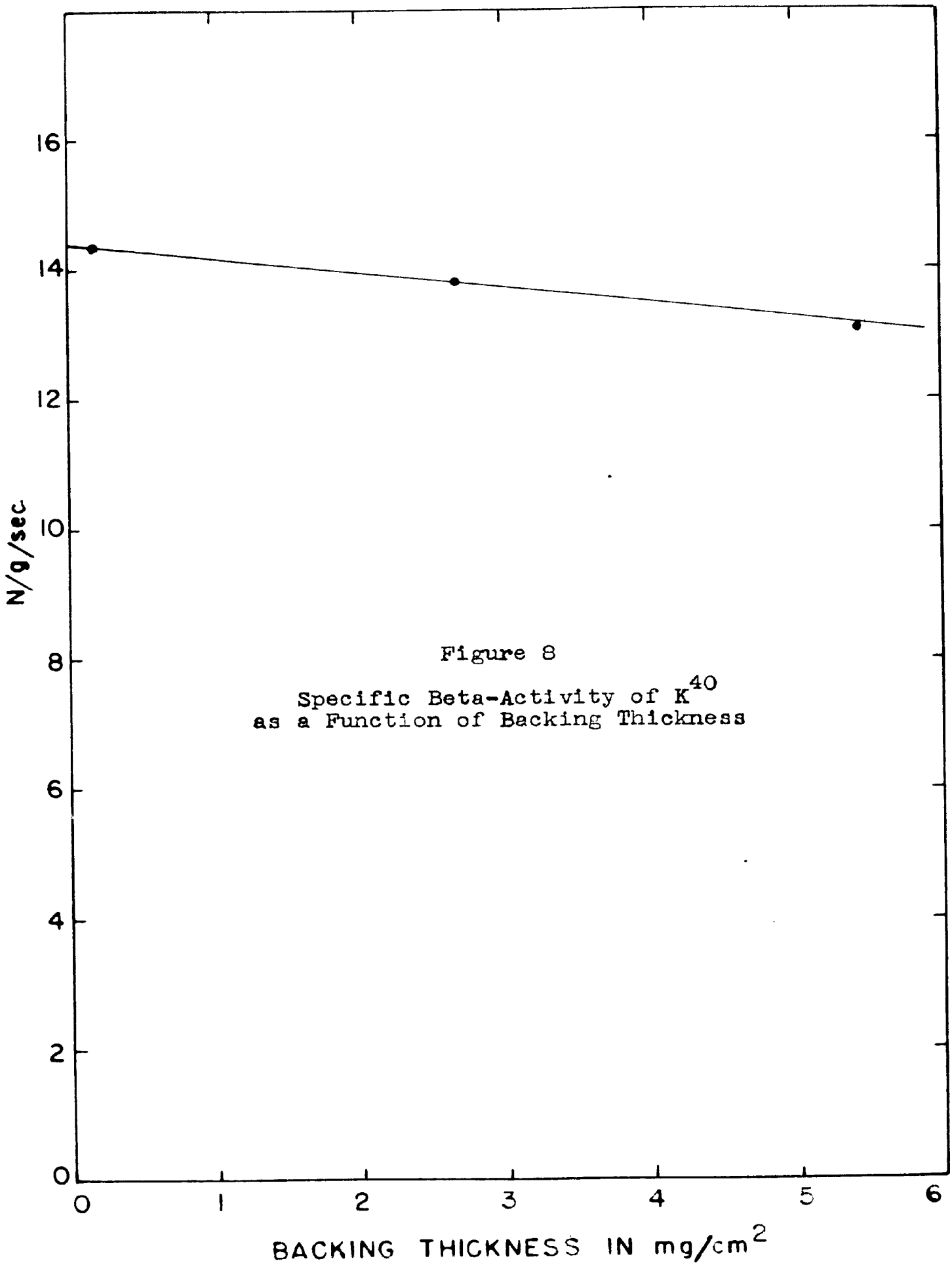


Figure 8

Specific Beta-Activity of K^{40}
as a Function of Backing Thickness

statistical counting errors, errors made in weighing the samples, and errors from the possible radioactive contamination of source- or background-foils. Except for sources less than 2 mg/cm^2 in thickness, the weighing error is small, being always less than 1 percent. Samples were weighed before and after counting, to insure that no material was lost. Usually, it was found that a small amount of KCl had been scraped off in handling, and if the loss exceeded 1 percent, the source data were discarded. Except for the thinnest sources, the statistical counting error also was 1 percent, or less.

The measured activity of 3 of the heavier sources was found to differ by about 5 percent from the expected values. The deviation in these cases was more than five times the statistical counting error, and the results from these sources were therefore discarded, it being assumed that in these cases, the sources had become contaminated or had been incorrectly weighed.

Measurements with all sources of less than 1 mg/cm^2 in thickness gave specific activity values that were consistently too high, by 5 to 15 percent, as compared with the other measurements. The activity of these sources varied from 1/5 to 1/2 of the background activity, and the probable errors became unduly large. It is not surprising, therefore, that the results obtained were not consistent with each other or with those with thicker sources. Although it is not clear why all these readings were high, the discrepancy is not regarded as serious. The measurements of these thin sources are not included in Fig. 7. A straight line may be drawn

through the plotted data, so that only one point falls more than 1 percent off the line. In view of this agreement, the statistical accuracy of the extrapolated value is better than 1 percent. Systematic errors, such as those caused by counter inefficiency, are probably not more than 2 or 3 percent, as explained above in the section on counter performance. The value for the beta-half-life is therefore given as:

$$T_{\frac{1}{2}\beta} = 14.2 \pm 0.5 \times 10^{8} \text{ years.}$$

6. 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 of Oak Ridge, and a source was made from this sample by evaporating the KCl, in a vacuum, from a tungsten filament onto a zapon film. Zapon is a commercial lacquer in an organic solvent. Very strong films, less than a micron thick, may be formed by putting a drop of the lacquer on a water surface. The lacquer spreads out over the water in a thin film, and, when dry, can be picked up on a wire frame. The film used in this experiment was built up by adding successive layers of zapon until it was about 1 micron thick, or about 0.1 mg/cm^2. A thin layer of silver (about 10μ g/cm^2) was evaporated onto the film to make it electrically conducting and the enriched KCl was evaporated on top of the silver to a thickness of 0.1 mg/cm^2. As before, the amount of KCl deposited was determined by weighing. The source was placed in the sliding-tray counter, with a blank zapon film for background measurement, and the counting rate was determined. The specific activity measured for this source

was 553 disintegrations/g KCl/sec. A small correction must be made to this value for the number of Auger electrons that are counted. It will be explained in Chapter IV that the number of counts to be expected from this source as a result of electron-capture decay is about 10/g KCl/sec, and this must be subtracted from the measured activity, leaving 543 for the specific activity of the source due to beta-rays. This corresponds to a half-life for beta-disintegration of 13.0×10^8 years. The uncertainty in this value is due largely to the probable error in the value for the isotopic abundance, ± 5 percent, and to the error caused by uncertainty in the number of electron-capture events counted, which is about 2 percent. The probable error in weighing the sample, due to inaccuracies in the balance, and the statistical errors in counting are each about 1 percent. The total probable error, therefore, is very nearly ± 5 percent, and the value for the half-life is:

$$T_{\frac{1}{2}\beta} = 13.0 \pm 0.7 \times 10^8 \text{ years.}$$

This value does not agree with the measurements made with ordinary KCl, within the probable errors. Since only a single measurement was made and the uncertainties are greater, this value is less reliable than the measurements with ordinary KCl sources. This value was, therefore, not used, and the half-life was taken as 14.2×10^8 years, as measured with the ordinary KCl sources.

7. Absorption Curve of K^{40} Beta-Rays

The absorption curve of the K^{40} beta-radiation was measured by placing successively heavier aluminum foils over an

ordinary KCl source of 13.5/mg/cm^2 and measuring the counting rates in one section of the sliding-tray counter. This absorption curve is the solid curve shown in Fig. 9. It was noted that the shape of the absorption curve was quite different from those measured by Hirzel and Waffler[30] and by Floyd and Borst,[21,23] their curves being concave toward the origin. It was suspected that the difference lay in the geometrical arrangement used. Therefore, an arrangement was tried with the source placed outside the counter, facing a mica window in the cover plate. The curve taken under these conditions was concave toward the origin and noticeably different from the previous curve. This curve is shown as the dotted curve in Fig. 9. As a further check on the influence of geometrical arrangement on the shape of absorption curves, the absorption curve of a P^{32} source was taken with a conventional, end-window counter, under three different geometrical conditions; (A) source and absorber about 1/2 cm from the counter; (B) source 10 cm from the counter but absorber next to the counter; (C) source and absorber 10 cm from the counter. The three resulting curves are shown in Fig. 10. In case C, when the absorber was placed in front of the source, the counting rate actually showed an initial rise because of the scattering toward the counter of particles originally directed away from it.

A further incidental experiment was performed to obtain an estimate of the back-scattering of radiation from the source backing. A source of P^{32} on 0.2 mg/cm^2 nylon was placed in front of an ordinary end-window counter, and as successively greater thicknesses of aluminum were placed behind

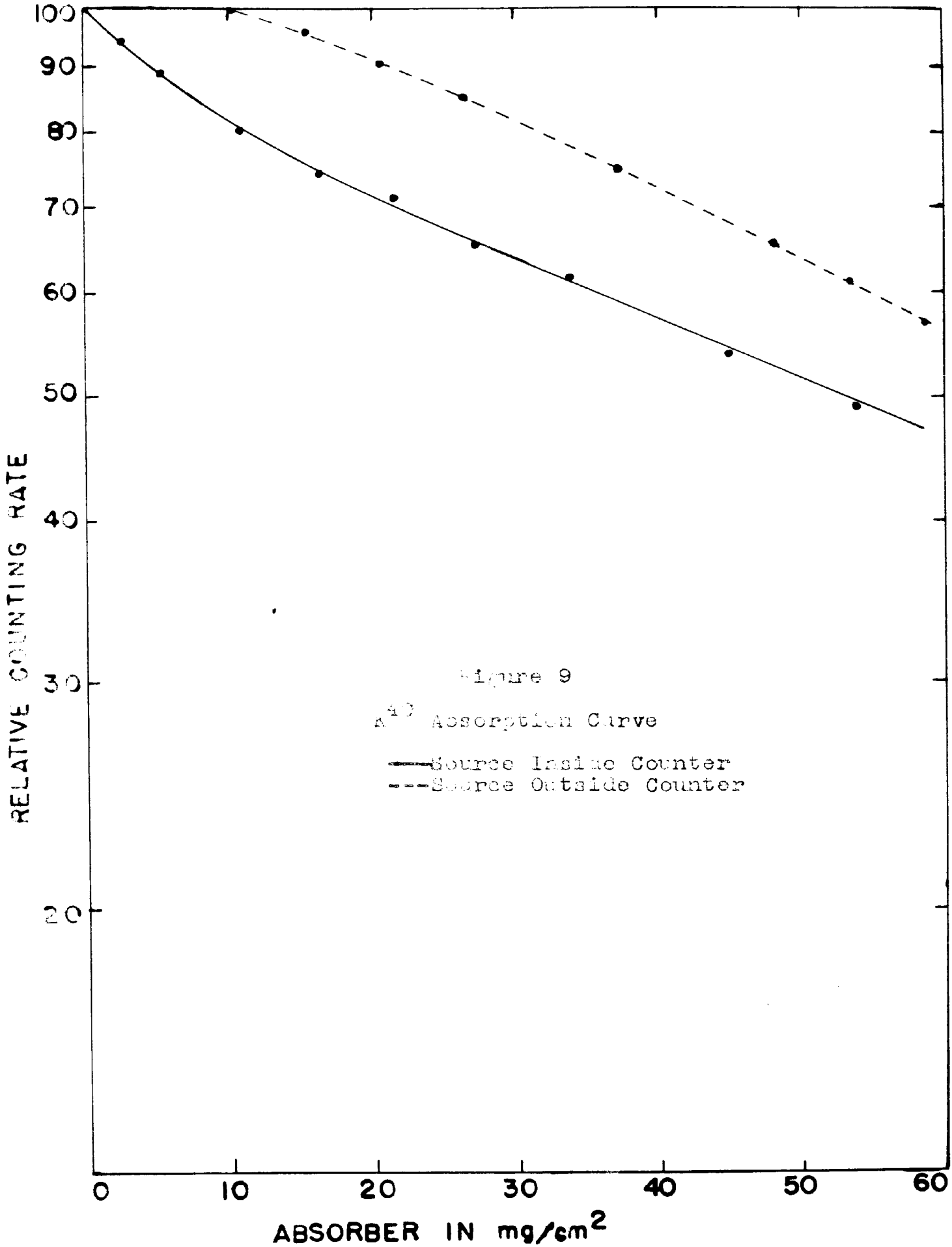
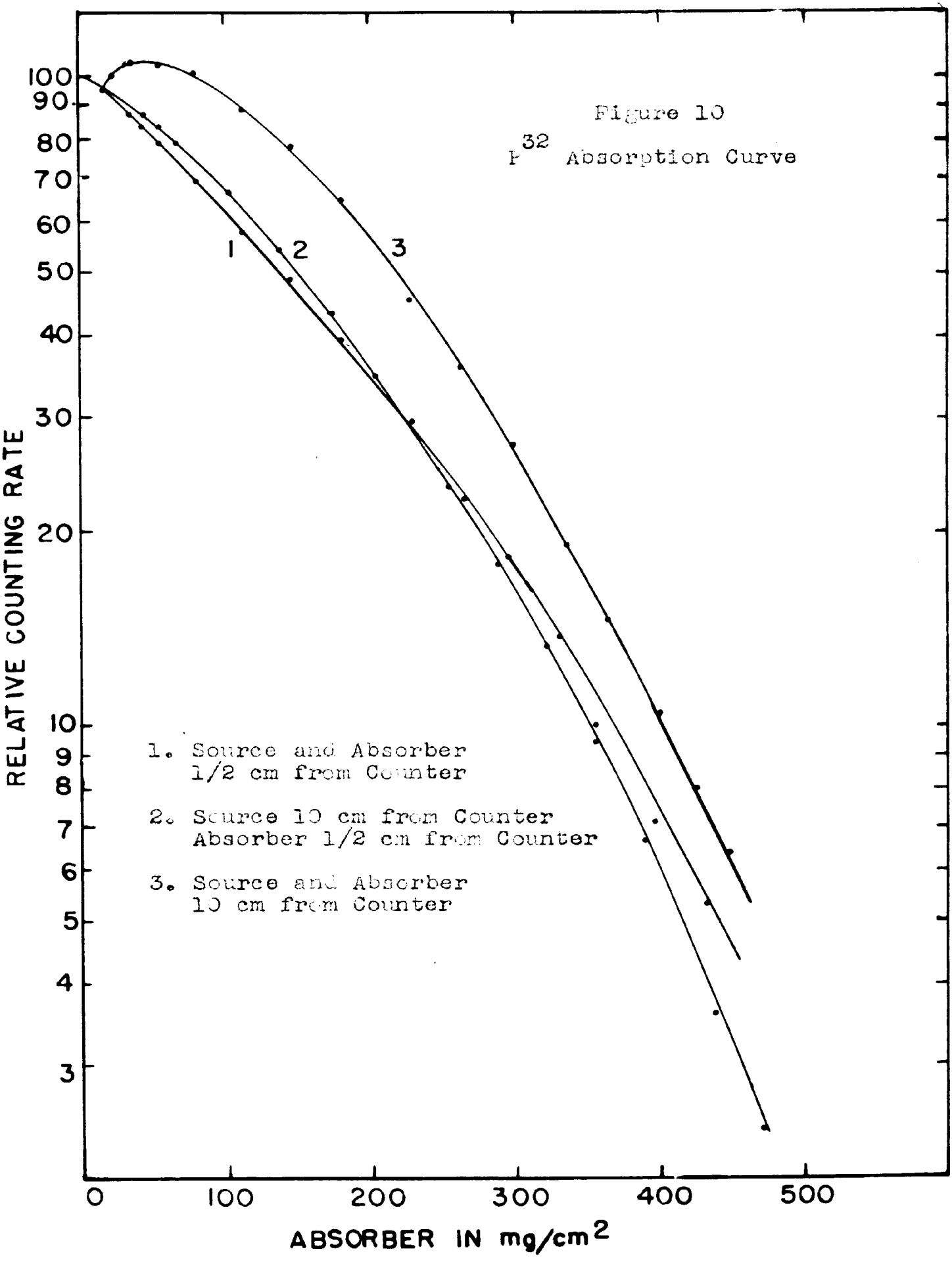


Figure 9

A^{40} Absorption Curve

——Source Inside Counter
---Source Outside Counter


Figure 10
P32 Absorption Curve
RELATIVE COUNTING RATE
100
90
80
70
60
50
40
30
20
10
9
8
7
6
5
4
3
1
2
3
1. Source and Absorber 1/2 cm from Counter
2. Source 10 cm from Counter Absorber 1/2 cm from Counter
3. Source and Absorber 10 cm from Counter
0
100
200
300
400
500
ABSORBER IN mg/cm2

the source, the counting rate was observed. The results showed a 15 percent increase in the counting rate with thick backing, as can be seen in Fig. 11.

On the basis of these data, it is possible to propose a probable explanation of the unique shape of the absorption curve of K^{40} taken with the source inside the counter: particles passing down through the backing are scattered upward by the bottom cover plate of the counter, which, in the experiment described, was about 1/4 in. below the source. Low-energy beta-rays do not have sufficient range to pass down through the source to be scattered, and then upward through the backing to be counted. Thus the high-energy part of the spectrum is augmented and there appear to be fewer low-energy particles, as is indicated by the curve (Fig. 9).

It was concluded that the shape of beta-absorption curves depends, to a great degree, on the conditions of measurement. No attempt was made to find the maximum range of the K^{40} beta-rays from the absorption curve, because of the shape dependence found and because the source activity was too weak to permit carrying the measurement below 10 percent of the initial activity.

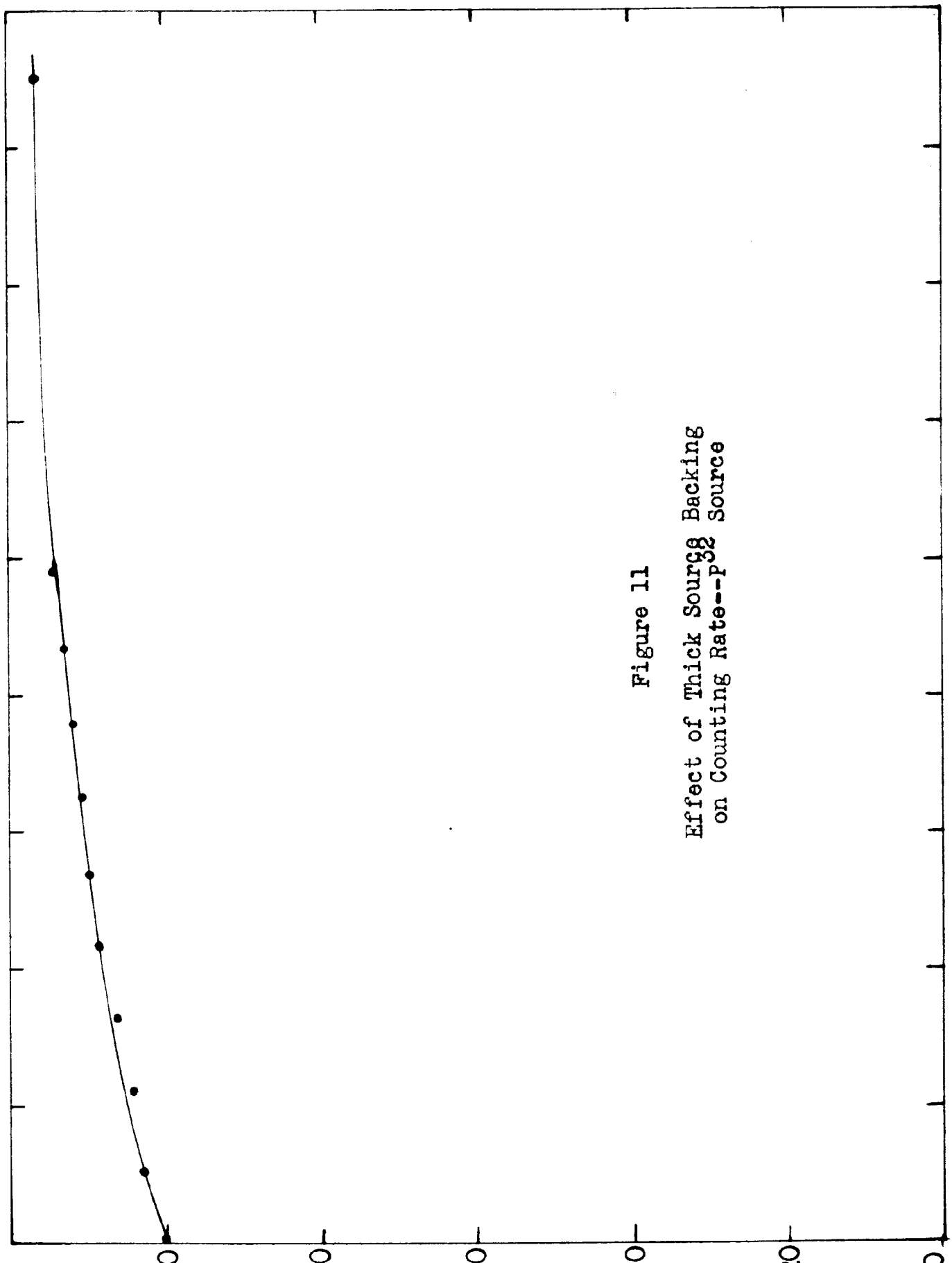


Figure 11

Effect of Thick Source Backing on Counting Rate--P^{32} Source

CHAPTER III

GAMMA-RAY MEASUREMENTS

The principal difficulty in making precise measurements of the gamma-ray intensity of K^{40} has been the lack of efficient gamma-ray detectors. For detecting high-energy gamma-rays, ordinary Geiger counters are less than 1 percent efficient. Scintillation counters, recently developed, have gamma-ray-detection efficiencies up to 30 percent, and for the measurment of the activity of these rays they were the counters used in the experiments described in this paper. Nevertheless, even when these more efficient detectors were used, massive sources of potassium were needed to obtain counting rates that were measurable above the background counting rate.

1. The Gamma-Counter

The source used with the scintillation counter consisted of 4 lbs. of KCl in a hollow cylindrical shell of brass, with a length of 6 in., an outside diameter of 6 in., and an inside diameter of 2 in. The walls of the brass shell were 1/16 in. thick, which was adequate to stop all beta-rays. In the center of this cylinder was placed the gamma-ray detector, a crystal of stilbene, 1 in. in diameter and 2 in. long. A lucite rod was provided to pipe the light scintillations from the Stilbene crystal to two adjacent 1P28 photomultiplier tubes (Fig. 12).

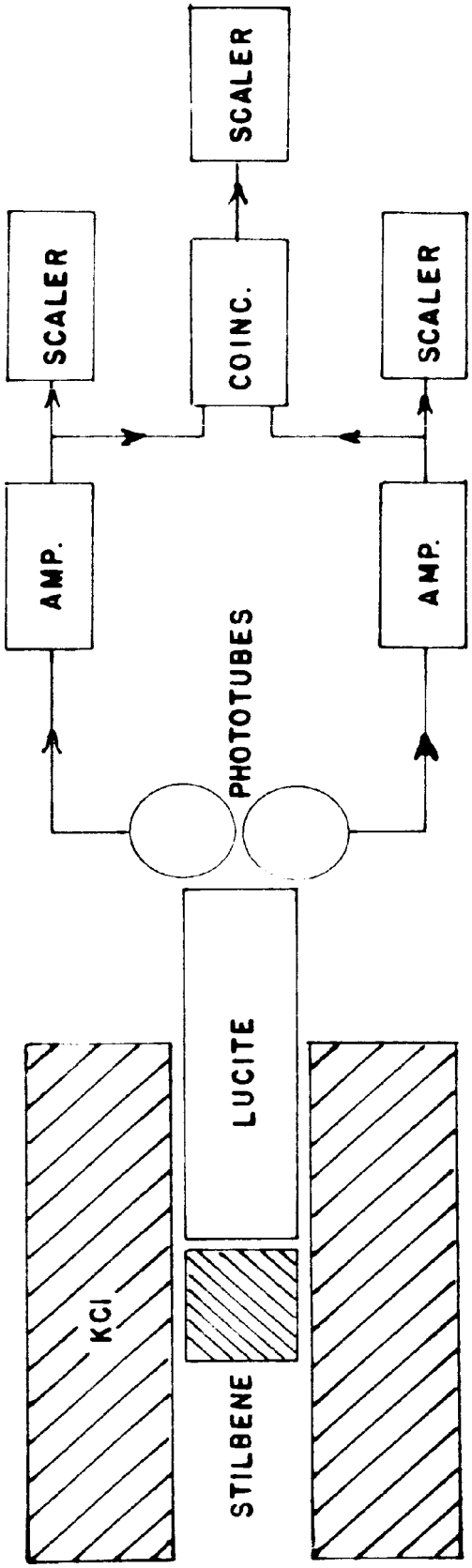


Figure 12

Gamma-Counter

Only the coincidences between the two photomultipliers were measured, and so, since the thermal-noise pulses in the photomultipliers, being random do not cause coincidences, it was possible to avoid the high thermal-noise background that is characteristic of a single photomultiplier tube. However, while the background was thus reduced from about 10 counts a second to 0.25 counts a second, the gamma-ray-detection efficiency of the coincidence counter was, unfortunately, only about 1/3 of the efficiency which could be obtained with a single photomultiplier. The reason for the loss in efficiency is simply that, in a large number of cases, only one of the two photomultipliers received enough light from the phosphor to record a count. In spite of this drawback, the signal-to-noise ratio obtained by the coincidence method was about 15 times as great as the ratio obtained with a single counter, and for this reason, the use of the coincidence method was well worth while. With this arrangement, the real counting rate from the K^{40} sample was about twice the background counting rate.

The electronic circuits used for recording the coincidence counts will be described only briefly, since they have been discussed in detail by J. R. Beyster.[44] The pulse output of each of the two photomultipliers is fed into a separate linear amplifier and pulse-height discriminator (Atomic Instrument Co. model 201), as shown schematically in Fig. 12. The discriminator rejects all input pulses below a predetermined size and generates an output pulse of constant magnitude (5 volts) for all input pulses of greater than the

predetermined size. In practice, the discrimination level is set for optimum signal-to-noise ratio. The discriminated output pulse from each photomultiplier is fed to a separate scaler and also to a common coincidence circuit. In the coincidence circuit, the pulse from each of the photomultipliers is fed into its own pulse transformer; the secondaries of the two pulse transformers are connected in series so that when pulses occur on them simultaneously, twice the output voltage of a single pulse is produced. The pulse-transformer output is then fed into another discriminator which selects the double pulses and rejects the single pulses. The resolving time of this coincidence circuit is 0.37 microseconds.

2. Method of Measurement

The determination of the absolute activity of the K^{40} sample was accomplished by a comparison method, which eliminated the need for direct measurement of the counter efficiency, the solid angle subtended by the counter, and the absorption of the gamma-rays in the thick source. K^{42} is known to emit a single gamma-ray in 25 percent of its disintegrations,[45] and the energy of this gamma-ray is 1.51 Mev, which is almost exactly the same as the gamma-ray energy of K^{40}; therefore, the counter efficiency and absorption coefficient are the same for the K^{42} and K^{40} gamma-rays, and K^{42} makes an excellent comparison source. Accordingly, a small amount of K^{42}, in the form of the chloride, is mixed into the KCl forming the K^{40} source so that the K^{42} is evenly distributed throughout, and the geometrical conditions are the same for it as for the K^{40}. Then the counting rates, with and without the K^{42}, are deter-

mined. From the ratio of the K^{42} to the K^{40} counts and the known activity of the added K^{42}, the activity of K^{40} can be calculated:

$$K^{40} \text{ activity} = \frac{(K^{40} \text{ counts})}{(K^{42} \text{ counts})} \times (K^{42} \text{ activity}) \qquad (9)$$

The K^{42} used for comparison in these experiments was obtained from the Carbide and Carbon Chemicals Co., Oak Ridge, Tennessee. The specific activity of the K^{42} was determined by counting the beta-rays from point-sources of various weights, mounted on nylon backing of .2 mg/cm^2 thickness. The position of the sources with respect to the beta-counter was rigidly fixed, so that the solid angle subtended by the counter was 0.974 percent. A supplementary experiment, in which the absorption curve of the K^{42} beta-radiation was taken, showed that the absorption of the beta-rays in the counter window and in the source itself was less than 1 percent and that it could, therefore, be neglected. The specific activities measured for the five samples are given in Table 7. Correction was always made, of course, for the decay of K^{42}, which has a 12.4 hour half-life. The decay of a sample of the K^{42} was followed through 6 half-lives, and the fact that the deviation from the expected decay curve was less than 1 percent indicated that there was no significant radioactive impurity in the sample. The negative results, also, of attempts to detect gamma-gamma coincidences indicated the absence of Na^{24}, which might be expected as an impurity.

3. Results

Five progressively larger amounts of the K^{42} were

TABLE 7

SOURCE-STRENGTH MEASUREMENT OF K^{42}

Source weight in mg.	Disintegration/gm/sec x 10^{-6}
2.30	1.25
2.30	1.17
3.20	1.19
3.55	1.17
4.95	1.21

TABLE 8

COUNTING RATE OF K^{42} MIXED IN K^{40} SOURCE

K^{42} in mg. added to K^{40}	Counts/gm/sec
62	43.0
214	46.2
382	47.2
712	45.6
1262	46.2

thoroughly mixed into the 4 lb. K^{40} source and, in each case, the resulting counting rates were measured with the coincidence scintillation counter. The data obtained for the specific counting rate of the K^{42} are given in Table 8. The specific gamma-activity of ordinary potassium calculated from these data is 3.6 gamma-rays/g K/sec., or about 12.7 gamma-rays/100 beta-rays.

The statistical errors in the experiment are due less to the statistical counting errors than to the errors in weighing the K^{42} samples and to the uncertainties in the uniformity of the mixture of the K^{42} into the 4 lb. KCl source. These errors may be evaluated from the agreement between the individual measurements of Tables 7 and 8. The maximum deviation from the mean in these tables is 4 percent and 6 percent, respectively, while the probable error is about 1 percent in each case. Thus the probable errors in determining the K^{42} activity and the K^{42} counting rate when it is mixed into the K^{40} source are each about 1 percent, and the probable error in the K^{40} counting rate is also about 1 percent, so that the total probable error (root-mean-square) in the K^{40} gamma-activity measurement is about 1.7 percent. There are also certain systematic errors which cannot be evaluated accurately. For example, the uncertainty in the solid angle subtended by the beta-counter in the K^{42} source-strength measurement may be several percent, and the K^{42} branching ratio measurement[45] may be in error and these would introduce a corresponding systematic error in the final result for the K^{40} gamma-activity.

In view of these uncertainties an allowance of about 8 percent probable error was made, and the gamma activity is given as:

3.6 ± 0.3 gamma-rays/gm K/sec.

CHAPTER IV

MEASUREMENTS OF ELECTRON CAPTURE

K^{40} is known to decay by electron capture, as well as by beta- and gamma-ray emission. In the radioactive decay of a nucleus by electron capture, the nucleus captures one of its own orbital electrons, usually from the innermost, or K, shell, and it is postulated that in the process of this disintegration to the element of next lower atomic number, the energy released is carried off by a neutrino.[36] Since the emitted neutrino cannot be detected experimentally, the electron-capture process cannot be directly observed, but since the characteristic x-rays of the new element are emitted as the vacancy in its K shell is refilled, the K-electron-capture process can be positively identified through these x-rays.

However, when the atoms emitting these x-rays have a low atomic number, most of the x-rays are absorbed in the outer shells of the atoms, and there is an accompanying emission of electrons.[45] This phenomenon, called the Auger effect, is similar to the internal conversion of gamma-rays. In the case of argon, which is the decay product of the K^{40} electron-capture decay, 88 percent of the emitted radiation consists of Auger electrons,[46] the remaining 12 percent being x-rays.

It is often possible for a nucleus which decays by

electron-capture to decay also by positron emission. Exactly the same decay product results in either case, but the energy available for positron emission is less, by two electron masses, than that available for electron-capture.[47] No evidence for positron emission in K^{40}, however, has ever been found. In 1941, Bothe and Flammersfeld[48] searched for it with a beta-spectrograph and concluded that the positron-activity is less than 1 percent of the beta-activity. More recently, Bell and Cassidy[26] searched for annihilation radiation from K^{40} with negative results. They concluded that the number of positron emissions must be less than 1 percent of the number of electron-captures.

1. Previous Investigations of Electron-Capture

That the number of disintegrations by electron-capture in K^{40} has not been satisfactorily established by previous investigators was pointed out in Chapter I, and, indeed, it is an extremely difficult quantity to determine precisely. Most of the previous determinations were made by analyzing potassium-bearing minerals for argon content, all of which, it was assumed, had been derived from the decay of potassium, and then calculating the amount of potassium that had decayed. For such calculations, it is, of course, necessary to estimate the geological age of the mineral analyzed and to assume that no argon has escaped from it since its formation. The validity of these assumptions seems doubtful, particularly in view of the fact that the electron-capture activities reported by different observers using this method show great discrepancies (p11). Somewhat more reliable appears to be the method of

Aldrich and Nier,[35] who used a mass spectrograph to measure the ratio of A^{40} to A^{36} in argon derived from potassium-bearing minerals. The argon from these minerals was found to be enriched in A^{40}, as compared with present-day atmospheric argon, and it was assumed that this enrichment was due to the decay of K^{40} to A^{40}. In this experiment it was not necessary to assume that no argon had escaped from the minerals, but it was necessary to assume that such escape had been shared by the two isotopes in proportion to their abundance. This assumption would be invalid, however, if the loss was by diffusion of the argon out of the mineral, since the lighter isotope, A^{36}, would be expected to diffuse somewhat faster. It was further necessary to assume that present-day argon in the atmosphere has the same isotopic composition as the argon which was originally occluded in the mineral, and the validity of this assumption is not established. On the whole, it appears that geological determinations of the number of electron-captures in K^{40} are subject to too many uncertainties to be reliable.

The only electron-capture measurement by a more direct method was made by Bleuler and Gabriel,[28] who used a Geiger counter to count the x-rays from K^{40}. It was necessary to distinguish the x-ray counts from the beta-ray counts, which were also present, and the counter was therefore arranged so that a gaseous absorber could be introduced between the K^{40} source and the counter, to absorb the x-rays selectively. The procedure was to measure the change in counting rate when

the gaseous absorber was introduced. The cellophane walls of the absorber chamber were heavy enough to prevent any Auger electrons from reaching the counter, and so only the 12 percent of electron-capture decays in which x-rays were emitted were detected. Moreover, the efficiency of the Geiger counter for x-rays was low, so that few of them were counted. The result was that most of the counting rate was due to beta-rays, and the reduction in counting rate when the absorber was introduced was only about 2 percent, part of which was caused by the absorption of the beta-rays in the gaseous absorber. Bleuler and Gabriel made no allowance for the scattering of either the x-rays or the beta-rays in the gaseous absorber. This scattering may have caused a large part of the 2 percent change in their counting rate, and their failure to take it into account introduces a considerable uncertainly in their results.

2. Description of the Electron-Capture Counter

The chief problem in measuring the electron-capture intensity with a Geiger counter is to distinguish successfully between the x-ray or Auger-electron counts, which are a measure of the electron-capture, and the beta-ray counts, which also come from the K^{40}. The experiments described in this thesis were undertaken with the idea that the electron-capture decay in K^{40} could be detected more readily through the Auger electrons than through the x-rays (the Bleuler and Gabriel method) because: (1) 88 percent of the radiation following electron-capture is in the form of Auger electrons; (2) Geiger-counter efficiency for Auger electrons is 100

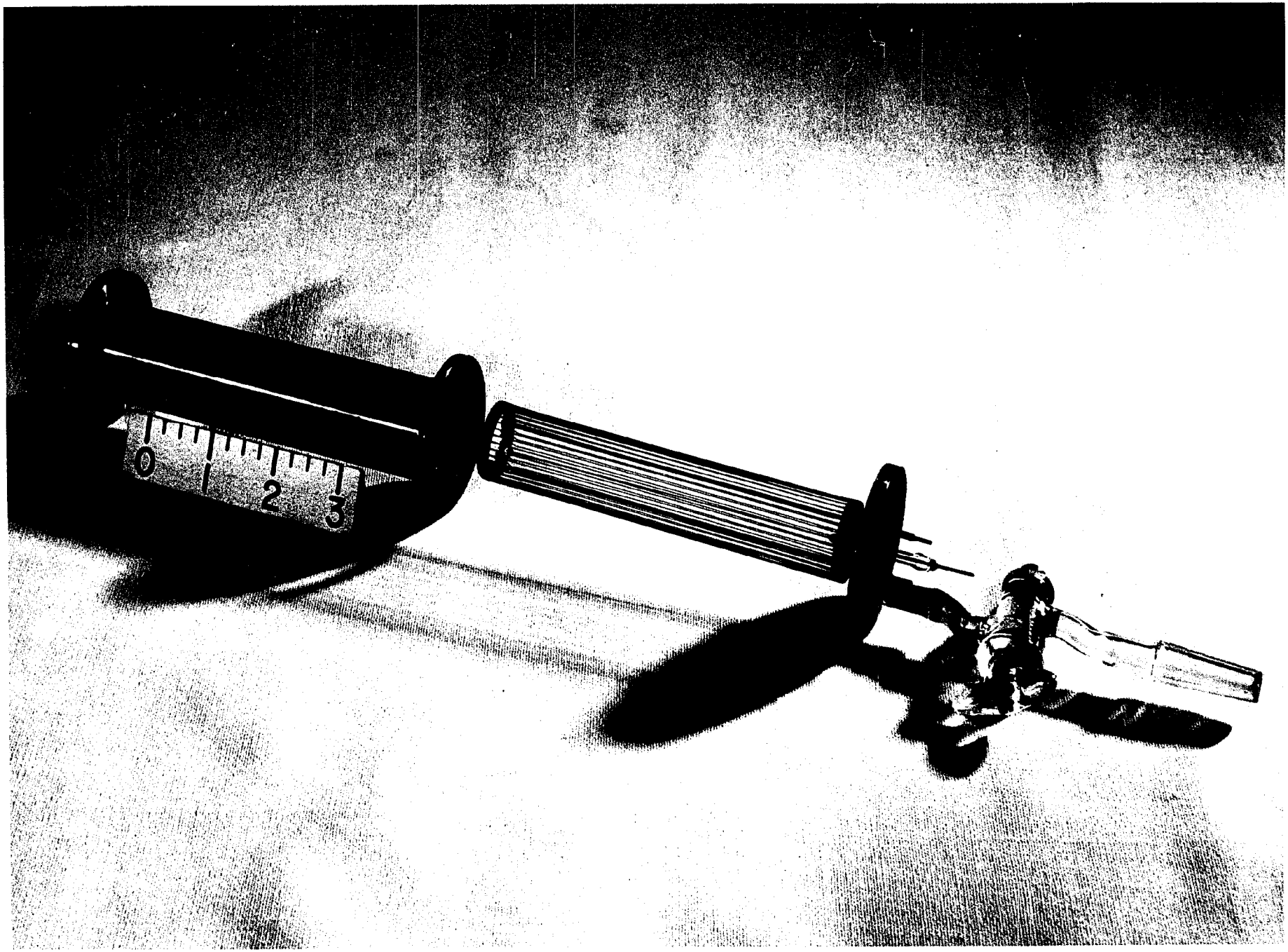
percent, while it is much lower for x-rays. There is, however, a difficulty in detecting the electron-capture through the Auger electrons in that these electrons have an energy of only 2.9 kev and their maximum range is, therefore, only about 60 $\mu g/cm^2$. The K^{40} source, then, must be made very thin, so that the Auger electrons can escape from it; consequently, only relatively small amounts of source material can be used, even though it is spread over considerable area. It is, therefore, important to keep the counter background as low as possible while still permitting the use of a source of large surface area. The problem is similar to that discussed in Chapter II, in relation to the beta-ray measurements, but is much more troublesome.

In order to provide a means for detecting the electron-capture process through the Auger electrons and for distinguishing the Auger electrons from the beta-rays, a special counter was designed. This counter is contained in a cylindrical steel shell, 1 1/4 in. in diameter and 6 in. long. The counter cathode is a cylincrical 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 counter is shown disassembled in Fig. 13. The special screen-cathode, which is the unusual feature of this counter, was made by winding a "squirrel cage", or cylindrical grid, of 10-mil copper wire, on a frame consisting of 2 copper rings, 1 in. in diameter, spaced 5 in. apart, and supported by tungsten rods sealed through the end plate of the steel shell. The

Figure 13

Electron-Capture Counter




0
1
2
3

10-mil wires forming the screen are spaced about 1/10 in. apart around the circumference of the supporting rings. When the area taken up by the tungsten support rods and the grid wires is calculated the transmission coefficient of the screen-cathode is found to be about 87 percent. A small bias voltage can be applied between the screen and the shell of the counter. The entire counter is shielded with 3 in. of lead to reduce the background.

The radioactive source was mounted in a thin layer on a rectangular, aluminum-foil blank of about 120 cm^2 area, and this blank was rolled up into a cylinder and slipped inside the steel shell of the counter. Thus, in effect, the source was mounted 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 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.

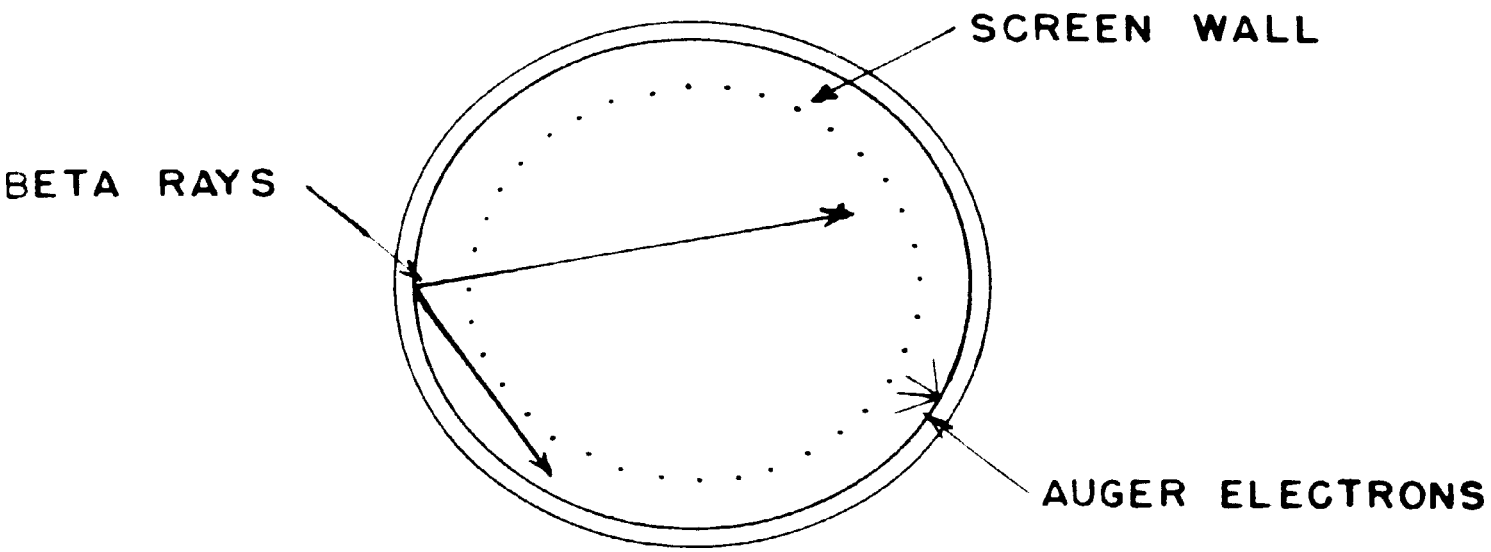
The small bias voltage applied (30 volts) has no effect on the trajectory of the high-energy beta-rays, and one might, at first thought, expect the beta-ray counting rate to be independent of it. However, applying the positive bias to the screen does increase the number of beta-ray counts, because some 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. 14. When the screen is made positive, the secondary electrons produced by the 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.

3. Counter Characteristics

To determine whether or not the counter had the expected characteristics described above, several tests were performed. Bias Curve. A source of Na^{24}, a beta-ray emitter, made in the University of Michigan cyclotron, was placed in the counter, and the counting rate, as a function of the bias voltage between shell and screen, was determined. Fig. 15 shows the bias curve, and it can be seen that at both positive and negative 30 volts 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

Figure 14

End View of Electron-Capture Counter



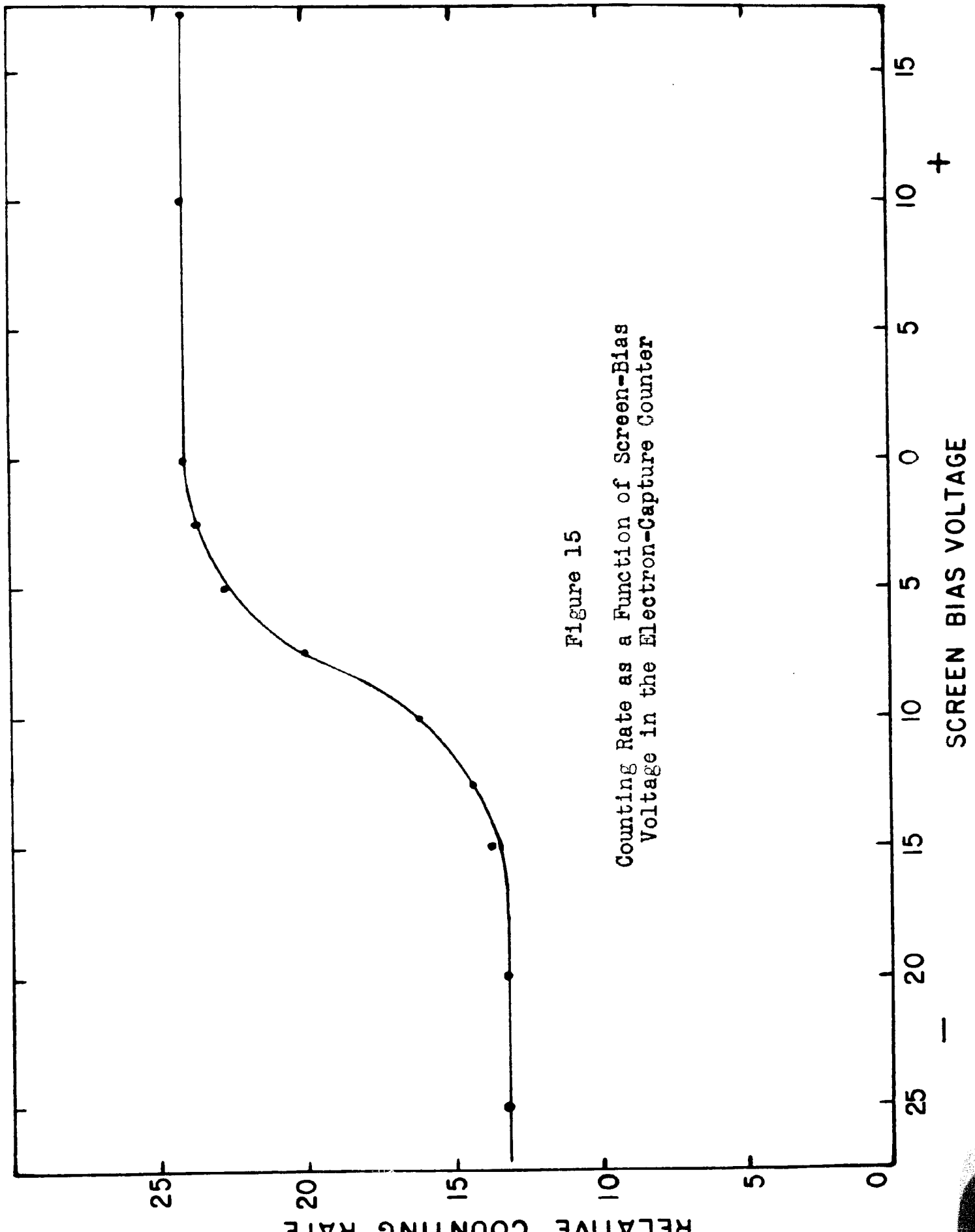


Figure 15

Counting Rate as a Function of Screen-Bias Voltage in the Electron-Capture Counter

negative 30 volts, all secondary electrons formed between shell and screen are prevented from entering the sensitive region of the counter, and that with the screen at positive 30 volts, all secondary electrons formed between shell and screen are attracted into the sensitive region and are therefore counted. There is no electron capture in the decay of Na^{24}, but the change in counting rate, as the bias was changed from positive to negative, was much greater than would be expected for a beta-emitter. Therefore, it seems probable that an isotope decaying by electron-capture, probably Zn^{65} occurring as contamination of the copper cyclotron target, was also present in the source. Its presence, of course, caused no difficulty in the determination of the bias curve.

Counter Characteristics for Beta-Rays. When the bias characteristics had been established, two beta-ray-emitting sources 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. As was explained above, an increase in counting rate was expected with the screen positive, since, in this case, secondary electrons from beta-rays not passing through the sensitive part of the counter are drawn into it. The sources used for this determination were Na^{24} and K^{42}, made in the cyclotron and sprayed onto aluminum-foil blanks to be placed in the counter. In the case of Na^{24}, the counting rate with the screen negative was found to be 71.9 $\pm$ 0.3 percent of the counting rate with the screen positive, or in other words, 71.9 percent of the

beta-rays pass into the sensitive part of the counter. In the case of K^{42}, the measured ratio was 70.0 ± 0.2 percent. Thus it was established that the ratio of screen-negative to screen-positive counting rates for beta-rays was about 71.0 percent and this would be the expected ratio in K^{40} if no electron-capture occurred.

Counter Characteristics for Auger Electrons. To determine the characteristics of the counter for Auger electrons, a source of Cr^{51} was used. Cr^{51} decays by electron capture, with 97 percent of the disintegrations going directly to the ground state of Va^{51};[48] the other 3 percent are by electron capture to excited states of the Va nucleus, with emission of gamma-rays as the Va falls to its ground state. About 75 percent of the x-rays from Va are converted to Auger electrons,[46] a smaller number than in the case of A because Va has a higher atomic number. The source of Cr^{51} was placed on an aluminum blank in the counter and the counting rates with positive and negative bias voltage determined, just as in the case of the beta-emitter measurements described above. The ratio of screen-negative to screen-positive counts was found to be 7 percent in this case -- 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 7 percent remaining here is attributed to conversion electrons resulting from the decay of Cr^{51} to an excited state of Va^{51}, with subsequent emission of converted gamma-rays, and to the x-rays emitted in 25 percent of the disintegrations, which are sometimes detected.

Range of Auger Electrons in the Counter. To establish beyond question 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 with the Cr^{51} source 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 of 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 negative-screen to positive-screen counts as a function of gas pressure is shown in Fig. 16. It is clear that 13 cm of gas pressure is sufficient to stop the Auger electrons from Va, 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.

4. Measurement of K^{40} Electron Capture

The determination of the electron-capture in K^{40} was then undertaken. An extremely thin source of KCl, enriched in K^{40} to isotopic abundance $0.40 \pm .02$ 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 1/6 the range of the Auger electrons from A. The real counts from this source, with positive bias voltage on the screen, were about .40 per second, or about half the background counting

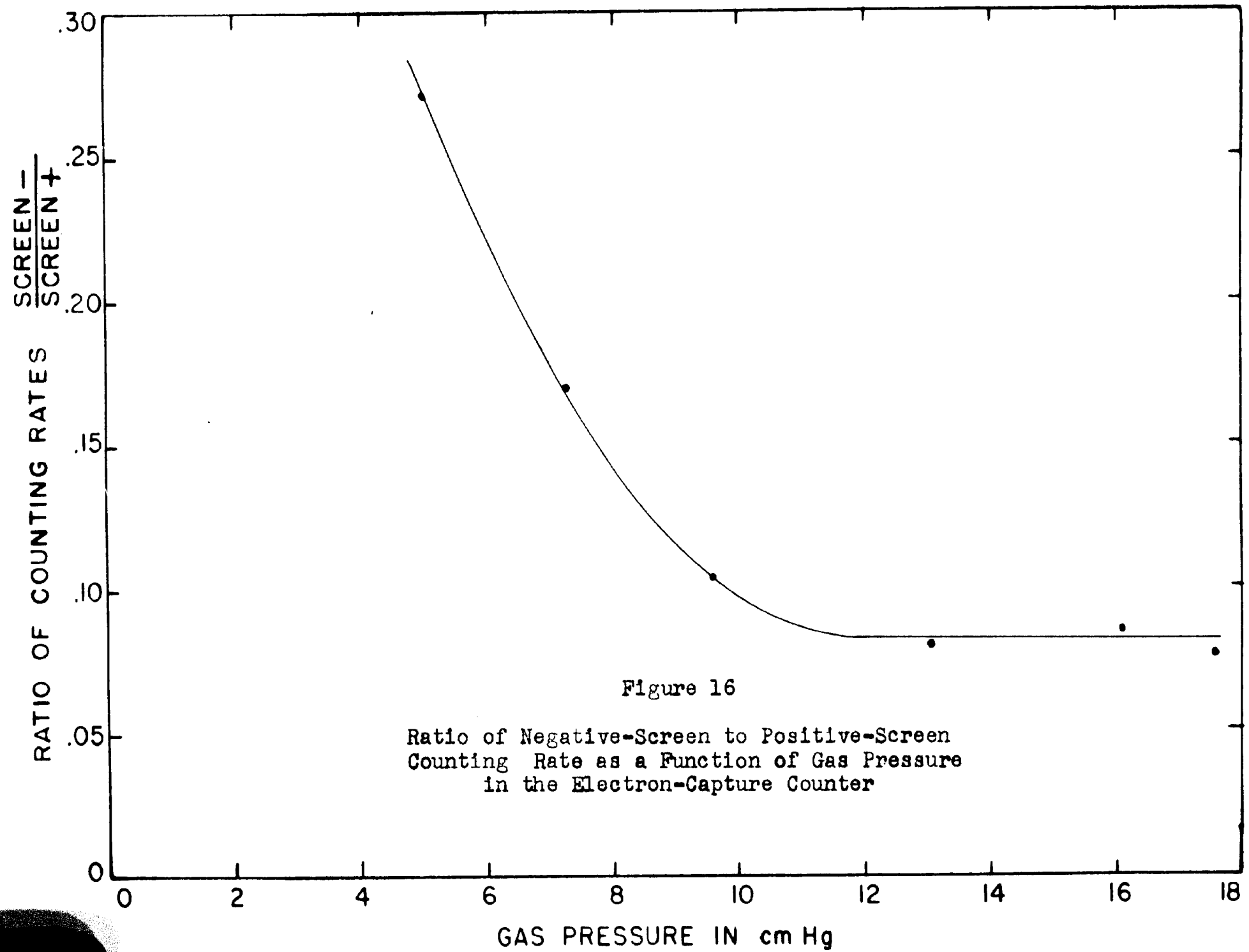


Figure 16

Ratio of Negative-Screen to Positive-Screen Counting Rate as a Function of Gas Pressure in the Electron-Capture Counter

rate. Several long runs were made with this source, alternated with runs in which the source was removed and the background measured. The several runs gave consistent values and the final ratio obtained for the negative-screen to positive-screen counting rates was 64.4 ± 0.2 percent. This ratio is to be compared with the values of 71.9 ± 0.3 percent and 70.0 ± 0.2 percent found for the two beta-ray emitters tried in the test described above. 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, and that, therefore, a small amount of electron-capture is taking place in the K^{40}. The number of decays by electron-capture were calculated by taking the counting rate obtained with the screen bias negative, which is due entirely to beta-rays, and dividing by the ratio of negative-screen to positive-screen counts for beta-rays (71 percent). This figure gave the number of counts due to beta-rays expected with positive-screen bias, which was subtracted from the observed positive-screen counts to obtain the excess due to Auger electrons. The calculated number of Auger electrons is 12/100 beta-rays. As was pointed out earlier, the number of electron-captures is about 12 percent larger than this, since in 12 percent of the electron-capture decays, x-rays, instead of Auger electrons, are emitted, and these are not detected. Accordingly, a value of about 13.5 electron-captures/100 beta-rays is obtained for K^{40}.

The error in this value is fairly large because of the low counting rates to be detected and the resulting poor

statistics, and it should not be regarded as exact; the error may be as much as 30 percent. It does show, however, that electron-capture actually takes place in K^{40}, and in about the right amount to permit the decay scheme of Fig. 18, with a gamma-ray in cascade with every electron-capture disintegration, there being 12.7 ± 1.1 gamma-rays/100 beta-rays. This decay scheme is proposed on the basis of the results in the present experiments. The value of Bleuler and Gabriel (190 electron-captures/100 beta-rays) is definitely ruled out, since such a large number of electron-captures would have meant a ratio of negative-screen to positive-screen counts of 30 percent in the present experiment.

5. Total Half-Life of K^{40}

When the decay constants of K^{40} for beta-decay (27.5 beta-rays/g K/sec.) and electron-capture (13.5 electron-captures/100 beta-rays) are combined, the total half-life of K^{40} is calculated to be $12.7 \pm 0.6 \times 10^{8}$ years.

6. Coincidence Experiments

There is, of course, a possibility that the gamma-ray in K^{40} is associated with the beta-decay, although this seems most improbable, as was pointed out in Chapter I (p. 4). However, an attempt was made, through coincidence experiments, to confirm the fact that the gamma-ray was in cascade with the electron-capture decay, rather than with a beta-decay. If the gamma-ray is in cascade with electron-capture, it should be possible to detect coincidences between the gamma-rays and the Auger electrons that result from electron-capture, unless the lifetime of the excited state of A^{40} is longer than 10^{-6}

seconds, corresponding to a large spin change in the gamma-transition.[50]

The counters used in this experiment were the sliding-tray beta-counter, described in Chapter II, and a six-section, multi-cell gamma-counter made up of sections, 6 in. in diameter and 1/2 in. thick, like those used in the sliding-tray counter, with 1/32 in. discs separating the sections. The multi-cell design was used to increase the gamma-ray efficiency of the counter, which was demonstrated to be about 6 percent. The characteristics of these counters have been described by J. R. Beyster,[40] and they will not be discussed here.

The source used was 10 mg of KCl, enriched in K^{40} to isotopic abundance 0.40 ± .02 percent. This source on a Zapon backing was one of the sources used in the beta-ray measurements with enriched sources described in Chapter II. The source was placed in the sliding-tray counter just as described there. Auger electrons from electron-capture, then, were able to escape from the source into the counter section it faced, but they were unable to penetrate the Zapon backing to enter the other section of the sliding-tray counter, since the Zapon thickness was about 0.1 mg/cm^2, or nearly twice the range of the 2.9 kev Auger electrons from K^{40}. Therefore, if the gamma-ray is in cascade with electron-capture decay of K^{40}, it should be possible to detect coincidences between the sliding-tray-counter section faced by the K^{40} source, but not coincidences between the gamma-counter and the other sections of the sliding-tray counter, where no Auger electrons are counted. The number of coincidences

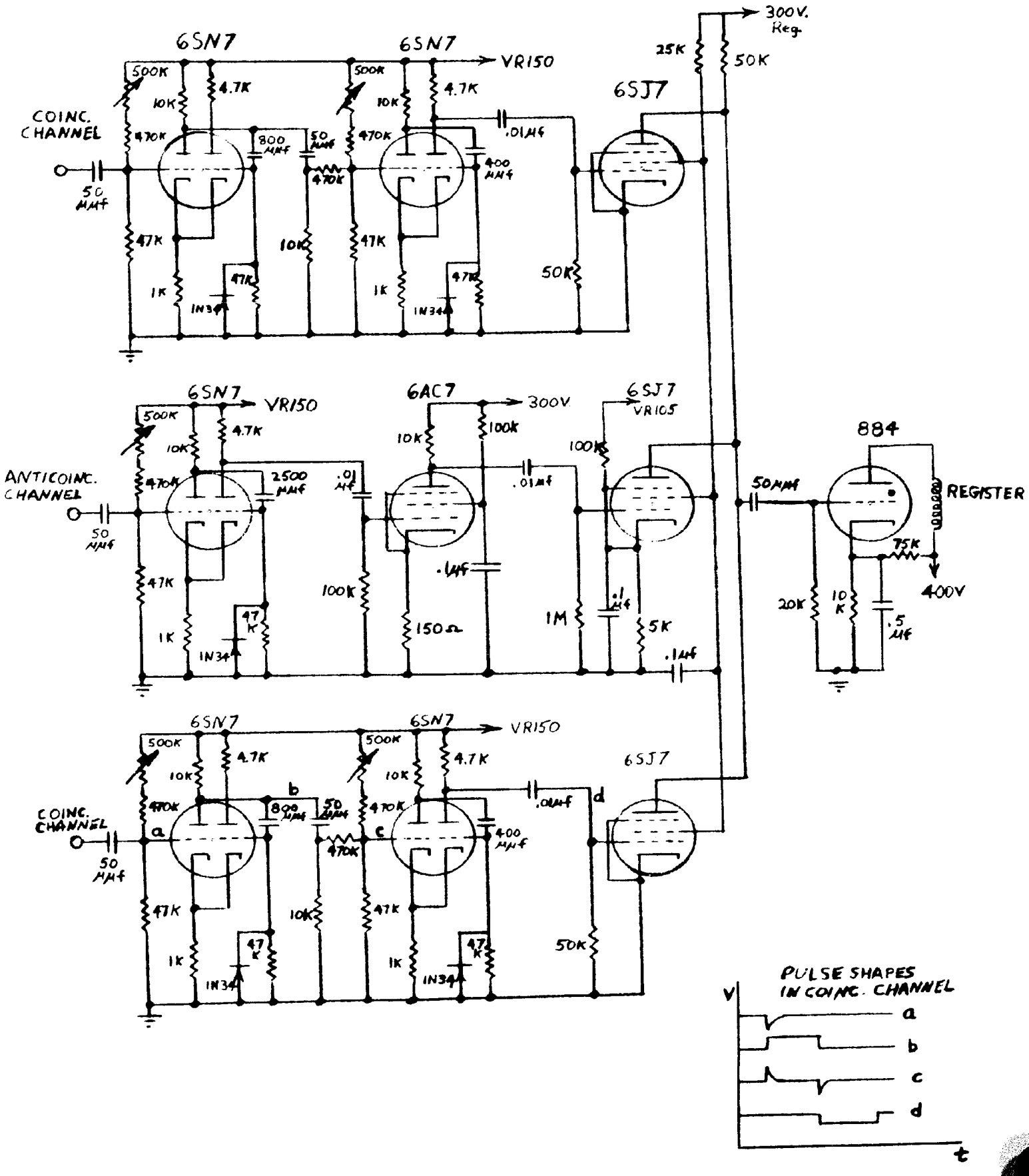
expected can easily be calculated. The 10 mg of enriched KCl has about 5 beta-decays per second and, therefore, about .60 electron captures per second. However, this source, which averages about 90μ gm/cm^2 in thickness, is thicker than the range of the Auger electrons, which is about 60μ gm/cm^2, and only about 1/3 of the Auger electrons directed into the section faced by the source can escape to be counted. Since only half the Auger electrons produced are directed toward this section, about .10 Auger electrons (.60 x 1/2 x 1/3) are counted per second. The gamma-ray counter, which is placed directly over the sliding-tray counter, subtends a solid angle of about 10 percent and has an efficiency for gamma-rays of about 5 percent. Therefore, only 0.5 percent of the gamma-rays coming from the K^{40} source in coincidence with Auger electrons will be detected, and the number of coincidences expected is about 1 per 2000 seconds.

When the coincidence experiment was performed, it was immediately discovered that high-energy cosmic rays were causing about 200 coincidences/1000 seconds and that it would be impossible to detect against this background the expected 1 coincidence/2000 seconds from K^{40}. To reduce the background, the first cell of the gamma-counter was placed in anti-coincidence with the sliding-tray counter and the remaining cells of the gamma-counter. A cosmic ray passing through the sliding-tray counter and the gamma-counter had to pass, also, through the anti-coincidence counter between them. Cosmic rays are strongly ionizing particles, and the counter efficiency for them is practically 100 percent. Accordingly,

nearly every cosmic ray will be counted in all three counters and will not be recorded as a coincidence. The counter efficiency for gamma-rays, on the other hand, is less than 1 percent, and a gamma-ray has a high probability of passing through the anti-coincidence counter without being counted. In that case, it may then be counted in the gamma-counter and register a coincidence. The anti-coincidence counter is therefore a means for rejecting cosmic-ray coincidences. It was possible, by this means, to reduce the cosmic ray coincidences from 200 to 10 per 1000 seconds. It was pointed out in Chapter II that the delays between the passage of a particle through one of these cell-type counters and the formation of the pulse on the anode wire are often several microseconds. To insure that every cosmic ray traversing the three counters was recorded as a coincidence, it was necessary to use a long resolving time in the coincidence circuit. It was necessary, also, to delay the pulses from the sliding-tray and gamma-counters, which were in coincidence, to prevent the recording of coincidences when these two counters fired relatively promptly but the anti-coincidence pulse was delayed; in events of this type, if the delay were not introduced, a coincidence would be recorded before the anticoincidence pulse came on. There are only an insignificant number of delays greater than 10 microseconds in these counters, and so the coincidence circuit was built with a 10 microsecond resolving time and a 10 microsecond delay in the two coincidence channels. The time delay and pulse lengthening required was accomplished with multivibrators. A partial-circuit diagram for the coincidence circuit is given in Fig. 17.

Figure 17

Coincidence Circuit



Attempts to reduce the coincidence background below 10 per 1000 seconds by the anticoincidence method were unsuccessful, and the origin of the remaining counts was not determined. They may have been caused by inefficiency of the anticoincidence counter, or they may represent cases in which the pulses were delayed more than 10 microseconds, making the anticoincidence feature ineffective.

In a final attempt to eliminate cosmic-ray coincidences, the coincidence apparatus was taken down into the salt mine of the International Salt Company, Detroit, Michigan. This mine is 1200 feet below ground level, and the cosmic-ray intensity in it is about one ten-thousandth the intensity at the surface. It was found that in the mine the coincidence background was about 1 count per 1000 seconds. That the coincidence rate was not reduced further is an indication that cosmic rays were not the only source of unwanted coincidence, since the accidental coincidence rate was only 3.4×10^{-3}/1000 seconds. Although this coincidence background was of the same order of magnitude as the number of real coincidences expected from the K^{40}, no significant number of coincidences could be detected above the background rate. However, since only 10 coincidences with an without the source were accumulated in the time available for the experiment, the statistics were inadequate and, therefore, the failure to detect real coincidences does not preclude their existence.

CHAPTER V

CONCLUSIONS

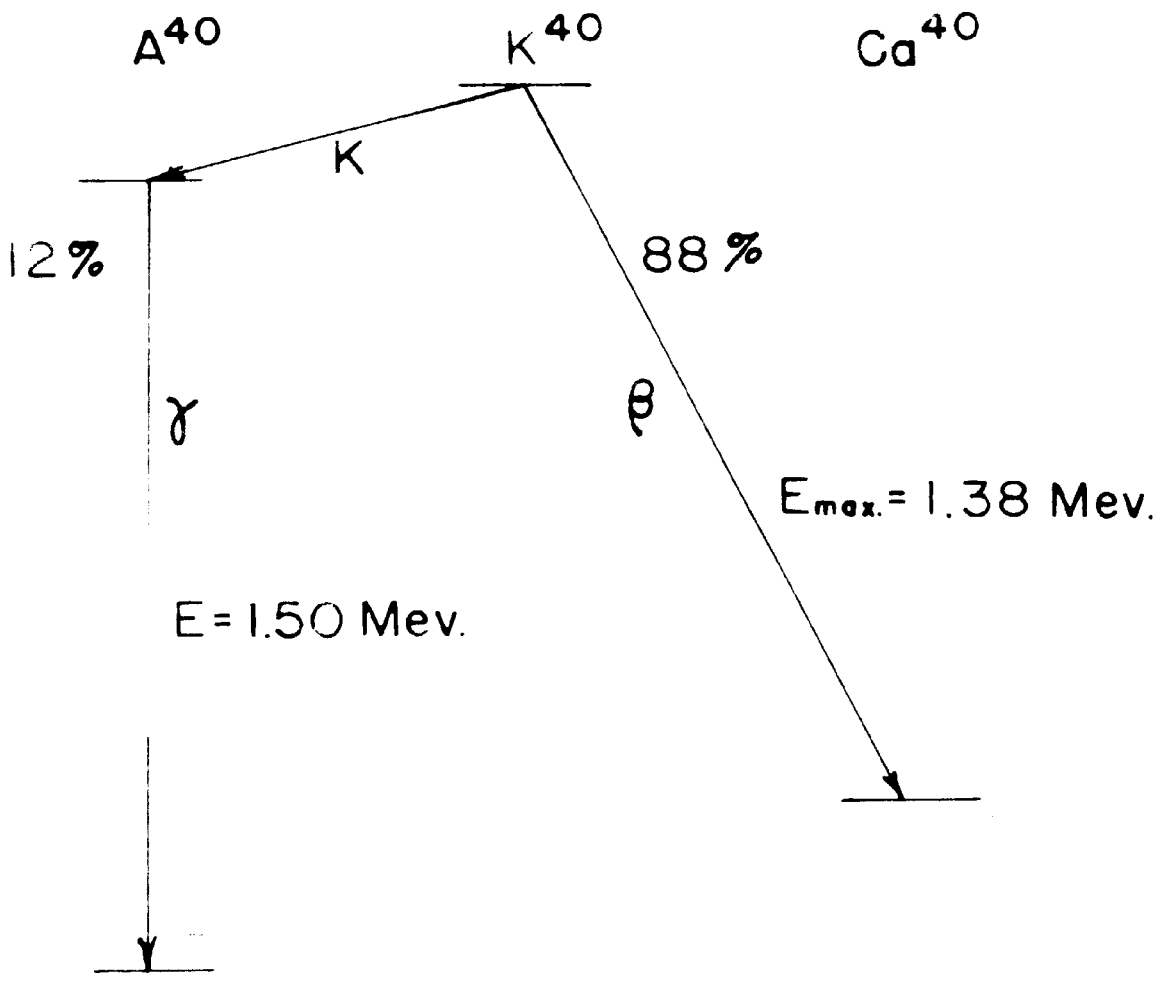
1. Results of the Present Experiments

In the present study of the disintegration processes of K^{40}, new determinations have been made of the beta- and gamma-ray intensities and of the rate of the electron-capture disintegrations. It has been found from these determinations, which are believed to be more accurate than earlier measurements, that the half-life for beta-decay in K^{40} is $14.2 \pm 0.5 \times 10^8$ years, based on the new observation of 28.3 disintegrations/g ordinary potassium/sec. For every 100 beta-ray emissions there were found to be 12.7 ± 1.1 gamma-rays and 13.5 ± 4.0 electron-capture disintegrations. New determinations of the energies of the beta- and gamma-rays were not undertaken since the best previous determinations seemed adequate to establish that the gamma-ray energy is greater than the maximum beta-ray energy.

On the basis of this fact and of the present measurements, a decay scheme for K^{40}, shown in Fig. 18, 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 assignment are the following: (1) The most reliable measurements of the beta-ray and gamma-ray energies, as discussed in Chapter I, indicate that the gamma-ray energy is greater than the maximum beta-ray energy. There are, from

Figure 18

Proposed Decay Scheme for K^{40}



the present measurements, approximately 9 beta-rays for each gamma-ray. If the gamma-rays were associated with the beta-ray disintegration, it would be necessary for part of the beta-decay to be to an excited state of Ca^{40}, followed by a gamma-ray disintegration to the normal 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, however, if no beta-rays have more energy than the gamma-rays. (2) The number of electron-capture disintegrations and gamma-rays measured were 13.5 ± 4.0 and 12.7 ± 1.1 per 100 beta-rays, respectively. The relatively close agreement between these numbers is probably fortuitous in view of the possible uncertainties in the electron-capture measurement. These numbers lend support, however, to the idea that there is 1 gamma-ray for each electron-capture disintegration, and that all the electron-capture disintegrations are to an excited state of A^{40}, with a gamma-ray following, as is shown in Fig. 18.

Since there are about 13.6 electron-captures per 100 beta-rays, approximately 88 percent of the K^{40} disintegrations are by beta-ray emission, and 12 percent are by electron-capture. From these figures it is calculated that the total half-life is $12.7 \pm 0.6 \times 10^8$ years.

2. Suggestions for Further Investigations

The decay-scheme of K^{40} could be determined more conclusively if coincidences between electron-capture disintegrations and gamma-rays could be detected. The attempts to detect such coincidences in this investigation were unsuccessa-

ful, as described in the previous chapter, but as sources enriched to greater abundance of K^{40} become available, and as more efficient gamma-ray counters with large sensitive areas are developed, coincidence experiments will become more feasible. At present, scintillation counters, which have the necessary efficiency, cannot be made with sensitive surfaces sufficiently large to make them useful in this experiment. There is every reason to expect improvement in this field.

Another question of interest is the shape of the beta-ray spectrum of K^{40}. Since the spin change of the nucleus in the beta-decay is probably 4, the spectrum should show a forbidden shape. The activity of K^{40} is too weak, however, to allow the use of conventional, magnetic-focussing spectrometers. One possibility for measuring the spectrum is to use a large ionization chamber at pressures high enough so that all beta-rays are stopped in the gas of the chamber. The size of the pulses from the ionization chamber would then be proportional to the amount of ionization produced by the beta-rays in being absorbed in the chamber, or proportional to the energy of the beta-rays. The spectrum could then be measured through the distribution of pulse heights from the ionization chamber. A pulse-height spectrometer, using a scintillation counter for the detector, has recently been used by P. R. Bell[24] to measure the K^{40} beta-spectrum. His preliminary results indicate that a third forbidden axial vector or tensor interaction produces a straight Fermi-plot above 700 kev.

Finally, it would be of interest to investigate the decay schemes of the naturally radioactive isotopes such as rubidium

and samarium, mentioned in Chapter I, about which very little is known.

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