

THE RADIOACTIVE ISOTOPE OF POTASSIUM

by

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Abstract

The isotopes of potassium were separated by means of a high intensity mass spectrometer and the beta-ray activity of each isotope was measured with a thin-walled Geiger-Müller counter. The measurements show that K^{40} is responsible for both of the known bands of beta-particles.

Introduction

The work to be described is part of a series of experiments initiated in 1924 by W. R. Smythe, who set out to separate the isotopes of solid elements by means of a high intensity magnetic lens type of mass spectrometer, with the object of collecting large enough samples to be used in certain experiments.

Much work has been done in many laboratories on the radioactivity of potassium since its discovery in 1905 by J. J. Thomson.¹⁾ Since the emission of beta-particles by such a light element is rather surprising, D. Dobrosserdow²⁾ and others have suggested that it must be due to the widespread existence of "eka-caesium", atomic number 87, as an impurity in ordinary potassium. Many attempts to remove this activity by chemical methods have failed, so this idea seems to have been generally abandoned. The discovery by Aston and Thomson³⁾ in 1921 that potassium has two isotopes, K³⁹ and K⁴¹, made it of interest to ascertain which of these is radioactive.

G. V. Hevesy and M. Logstrup⁴⁾ prepared, by fractional distillation, a specimen slightly heavier and slightly more radioactive than ordinary potassium. They concluded that the active isotope has a mass greater than 39. Elaborate experiments, carried out to find the end product, Ca³⁹ or Ca⁴¹, of this disintegration, were unsuccessful. This suggested the possibility that a rare isotope K⁴⁰ is responsible for the radioactivity, with the well known Ca⁴⁰ as the end product. Further measurements with a Geiger-Müller counter

by G. v. Hevesy, W. Seith, and M. Pahl⁵⁾ on the heavy samples of potassium showed an increase in the radioactivity of $4.43 \pm 0.53 \%$. A determination of the atomic weight by Hönigschmid and Goubeau⁶⁾ indicated an increase in the concentration of K^{41} of $4.2 \pm 0.84 \%$. They asserted that these results prove the radioactivity of potassium to be due to K^{41} and that they definitely exclude an hypothetical K^{40} . More precise measurements of the atomic weight of potassium⁷⁾ and of v. Hevesy's heavy potassium,⁸⁾ together with recent measurements of the abundance ratio^{9,10)} of K^{41} caused v. Hevesy to withdraw his earlier statement and his last conclusion is that K^{40} must be the radioactive isotope.¹¹⁾ Finally, it was discovered by Nier,⁹⁾ and confirmed by Brewer,¹⁰⁾ that K^{40} constitutes about 1 part in 8300 of ordinary potassium. Meanwhile, theoretical reasons, the most cogent of which has already been mentioned, had been advanced for the assignment of the radioactivity to K^{40} . Klempener¹²⁾ and Newman and Walke¹³⁾ considered that of the hypothetical group, K^{39} , K^{40} , K^{41} , K^{42} , the only one which could give the observed activity was K^{40} . After the discovery of K^{40} Sitte¹⁴⁾ pointed out that, since Ca^{40} and Ar^{40} are both well known stable atoms, if K^{40} atoms can disintegrate with the emission of electrons they should also disintegrate with the emission of positrons. Because careful investigations have failed to detect positrons he concludes that the experimental evidence is against the radioactivity of K^{40} , and suggests that some other unknown isotope, probably K^{43} , is responsible.

Clearly, only the determination of the radioactivity of each isotope, separately, can answer this question conclusively. The only known method of preparing a pure isotopic sample from a group of three or more is that of mass spectroscopy.

The spectrometer which has proved to be successful for the separation is one designed by W. R. S. and L. H. Rumbaugh, improved by S. S. West, and finally put into operating shape by the writer. The work of the aforementioned need only be summarized here.^{15,16,17)} From their experience with an earlier apparatus (designed and constructed by W. R. S.), W. R. S. and L. H. R. were able to design a very satisfactory instrument for this purpose. The spectrometer was constructed in the Astrophysics Machine Shop of the California Institute. L. H. R. set it up and made some measurements of the intensity and resolving power. S. S. W. devoted most of his energies to devising a satisfactory form of ion source, in which endeavor he was successful. He also tried a number of methods of collecting the separated isotopes, which problem proved to be more troublesome than was anticipated.

The situation in the fall of 1934 when the writer took over this work was as follows: the resolution of the instrument was just that predicted by theory, the intensity was too low to collect samples large enough to be of any use, and there was no satisfactory method of collecting the isotopes.

This thesis will be divided into two parts: the first part

will deal with the technique of separating the potassium isotopes, and the second part will be devoted to the measurement of their beta-ray activity.

Separation of the Isotopes of Potassium

Modifications of the ion source

The ion emitter used by L. H. R. and S. S. W. was a Kunsman catalyst. This is principally ferroso-ferric oxide containing approximately one per cent of potassium oxide. The oxide was ground to a powder, reduced in an atmosphere of hydrogen at 900°C., and was then ready for use. When the supply of this emitter (it was supplied by A. Keith Brewer of the Fixed Nitrogen Laboratories) was exhausted it was necessary to prepare the emitter here. This required a high temperature furnace and was hardly feasible with the facilities on hand. Upon the suggestion of W. R. S., the writer resorted to a different method of preparation; a mixture of ferric nitrate and potassium carbonate was heated in an evaporating dish until the ferric nitrate melted (at 47°C.). The potassium carbonate then dissolved in the liquid. The mixture was next heated to dryness. The solid mass was cracked into small pieces and heated in an iron crucible, driving off the oxides of nitrogen and leaving a red mixture of the oxides of iron and potassium. This solid mass was ground in a mortar, reduced in the same manner as the Kunsman catalyst, ground again, and was then ready for use. The ratio of the mass of ferric nitrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$) to potassium carbonate used was 6.15 to 1. This gives an emitter containing about thirty times more potassium than the Kunsman catalyst.

In the course of a series of experiments to determine the properties of various substances as emitters a very effective source of copper ions was found. An attempt was made to use a potassium ion source similar to that described in the last paragraph but with a base of copper instead of iron. Initially the emission was as great as with the iron base, but the emission lasted only half an hour. But at a temperature much lower (a dull red heat) than that at which potassium ions were emitted, copper ions were observed, singly charged. The temperature of emission was rather critical, but the emission was observed when this temperature was approached from either above or below.

The writer attempted to make a source of lithium ions similar to the iron base source for potassium, but without any success. It should also be mentioned here that spodumene (a lithium-aluminum silicate) showed a weak emission of lithium ions which lasted only a few minutes. At the highest temperature attainable with the best source spodumene showed no additional emission.

The upper limit of the temperature attainable with the first source of S. S. W. was set by the heaters, which were molybdenum helices insulated from the source block by Stupekoff tubes. Accordingly, he replaced the molybdenum wire by tungsten wire and the Stupekoff insulators by tubes of magnesium oxide, which will stand up at temperatures up to 2000°C. These insulators have the added advantage that if they are over-heated they do not melt and attack the heater, but volatilize instead. The magnesium oxide tubes were

prepared by baking a paste of fine sintered MgO wetted with a solution containing 2% of $MgCl_2$ and formed in a graphite mold, in an induction furnace.¹⁸⁾

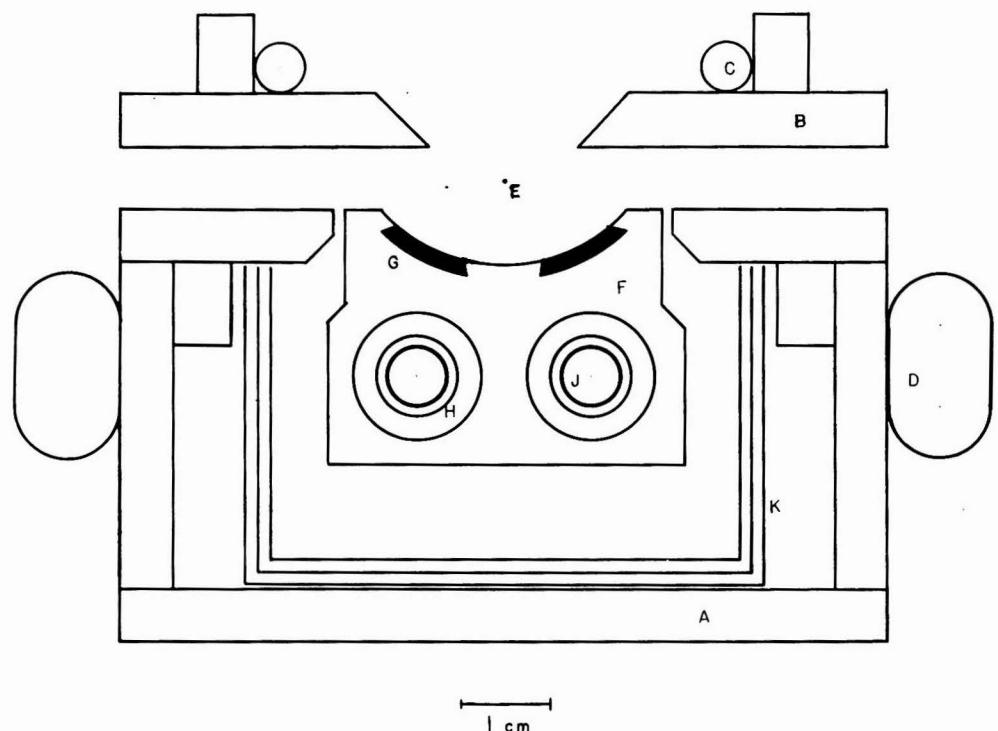
S. S. W. proposed also replacing the iron source block by one of graphite, which the writer did. Extensive tests were made with this source, with very discouraging results. A broad peak persisted in all experiments and it was never possible even to resolve the two main isotopes of potassium using this source. Finally, it appeared that the carbon source block was attacking the magnesium oxide insulators, causing a continuous evolution of carbon monoxide, the presence of which gas near the source, where there was a strong electric field, was responsible for the great width of the peak. This gas was not indicated by the Pirani vacuum gauge, but the outlet to the vacuum pump was near the source and the vacuum gauge was at a great distance from the source. The writer nickel-plated the source block, both on the inside of the cylindrical holes and on the outside of the block, but the nickel peeled off at high temperatures. At this point the carbon source block was abandoned and the old type of iron source block was again put into service.

Three polished nickel reflectors were placed around the source in order to minimize radiation losses.

Another slight modification was in the water cooling system for the copper box which housed the source: the 3.2 mm (inside diameter) copper tubing inside the box was removed (to make room

for the reflectors) and a 12.7 mm (inside diameter) copper tube, partially flattened, was bent around and soldered to the outside of the source box. A cross section of the final source assembly is shown in Fig. 1.

A significant advance was the adoption of a new method of packing the emitter in the grooves of the source block. The method that had previously been used with this type of source was to place the powdered emitter on the block and to press it into the shallow grooves with an iron cylinder which just fitted the surface of the block. At high temperatures the emitter would shrink and break up, leaving an irregular surface. Furthermore, the contact between the block and the emitter was so poor that heat was not conducted to the emitter at a sufficiently great rate, with the result that when the iron block was at a yellow heat the emitter still appeared black. The new method, suggested by W. R. S., was to clamp a cylinder over the source block and to pour small quantities of the emitter in one end of the groove (the other end being closed), packing each charge tight with a phosphor-bronze chisel. The two grooves packed in this manner were 25 cm long, 9.5 mm wide, and 1.6 mm deep. The emitter after it was packed had an excellent surface; it was hard and behaved almost like a solid metal surface. At high temperatures the emitter showed nearly the same color as the source block. Several hours are required for the preparation and assembly of the source. During this time considerable oxidation of the emitter



- A Copper source box
- B Copper slit plate
- C Water cooling tubes
- D Water cooling tubes
- E Tungsten focussing wire
- F Iron source block
- G Ion-emitting substance
- H Magnesium oxide insulating tubes
- J Helical tungsten heaters
- K Polished nickel reflectors

SECTION OF SOURCE ASSEMBLY

occurs. For this reason the source is again reduced in an atmosphere of hydrogen after it is assembled in the mass spectrometer, by admitting hydrogen to the system at 5 cm of mercury pressure and keeping the source at a dull red heat for 15 minutes, for which 2 kilowatts of power are required.

The total emission from this source is far greater than ever obtained before. In one run the total useful emission of K^{39} was 20.4 mg; when this run was discontinued the resolution was still good and there was no sign of exhaustion of the source.

The Collector

The first method, tried by S. S. W., of collecting the ions was to place a cup behind a slit at the focus of the spectrometer, which cup was at such a potential that ions striking it had only a few hundred volts velocity. In some cases a grid was placed across the slit to produce a field of suitable shape. One difficulty with this method is that the field is always of such a shape that the incoming ions tend to be deflected (the beam is 45° wide) in such a way that they never touch the collector.

The writer constructed a collector as shown in Fig. 2. Here there was no chance for the ions to be deflected, for the opening of the collector was kept at a low potential due to the proximity of the grounded copper strip. This collector behaved much as was expected: the atoms of the ion beam were deposited on the collector, concentrated at D and E, as was revealed by inspection of the collector after it was removed from the apparatus; the usual discoloration appeared where atoms had been deposited. But with this collector a new difficulty attracted our attention. Any incoming ions that struck the copper strip did so with a velocity of 5000 electron-volts. Secondary electrons were ejected from this strip, and the field inside the collector was of such a direction as to accelerate these electrons. The potassium deposit on the collector was cleaned off wherever these electrons struck.

At various times in the course of these experiments the

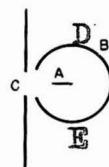
collector was cooled with liquid air. A reservoir for liquid air with a capacity of 30 cm³ and with provisions for filling it from outside the apparatus was installed in the collecting chamber, and the collector was mounted on the end of this tank, as is shown in Fig. 4. The greased conical joint B allowed the collector to be moved until it was in front of a window in the side of the collecting chamber, so that it was possible to see what was going on without opening the apparatus. It was hoped at first that liquid air cooling, without any retarding field, might be sufficient, but it was not, and it was soon obvious that a combination of liquid air cooling and any of the retarding fields tried previously would not work either.

There are difficulties inherent in the methods of retarding fields and liquid air cooling. The field must always be of such a direction as to accelerate any electrons that are free near the collector, and there will always be some secondary electrons where there is such a fast ion beam. These electrons will clean off any deposit that is formed. A collector at a very low temperature will act as a trap for any condensable vapors in the system. There is evidence that this actually happened, for when the collector was cooled an insulating layer was formed on its surface, and this layer was not penetrated by ions of several hundred volts energy.

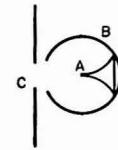
At this point it seemed advisable to try an entirely different method of collecting. We observed in some of the earlier experiments that if fast ions struck the collector they were reflected with

low energies. Relatively thick deposits of potassium that had been either reflected or evaporated from the collector were found on the back of the plate which held the focal plane slit. Accordingly a collector was constructed as shown in Fig. 3. The incoming ions struck the cylindrical wedge A and landed eventually on the concave walls of the outer cylinder. This method of collecting proved to be satisfactory. A silvery coating appeared on the inside of the collector after 0.1 mg (calculated from the collecting current) of potassium had been deposited; furthermore, the coating remained for several days in the vacuum. Frequently in earlier experiments apparent deposits of potassium disappeared after they stood for a few hours in the vacuum system. No liquid air cooling was necessary with this collector.

It developed in the course of later experiments that a beam of high energy ions impinging on a brass target has a rather drastic effect. The deposit of potassium formed on the collector appeared to be mixed with brass; the coating of brass was so thick that it peeled off, in much the same way as a poorly deposited evaporated film of metal peels. Also, the bevelled brass slit edges were attacked by the ion beam to such an extent that the 5 mm slit was widened to 6.5 mm. The brass was then inlaid with 0.15 mm molybdenum, and 4 mg of K^{39} ions drilled a hole through the molybdenum and 1 mm into the brass, as shown in Fig. 5A. Tungsten proved to be more resistant under ion bombardment: a hole 4 x 6 mm in area and with a



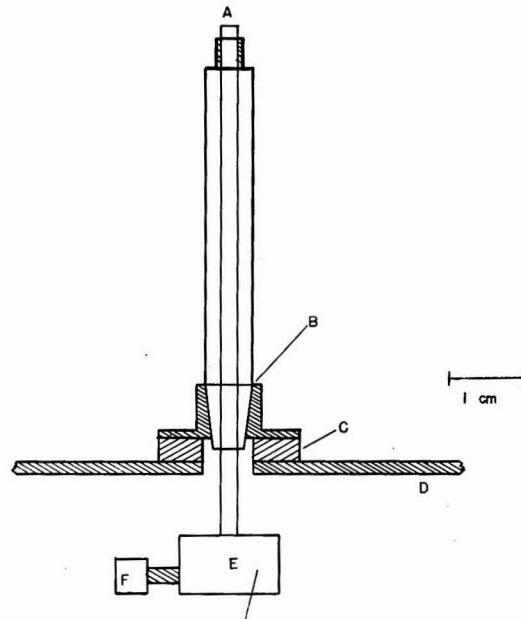
A Copper strip at ground potential
 B Brass collector at high positive potential
 C Slit at focus of spectrometer



A Wedge-shaped brass target
 B Brass collector
 C Slit at focus of spectrometer

FIGURE 2 . SECTION OF COLLECTOR

FIGURE 3 SECTION OF COLLECTOR



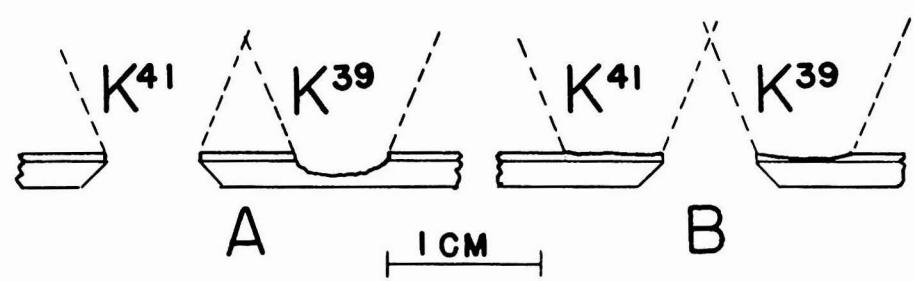
A Liquid air filling tube
 B Greased conical joint
 C Redmanol insulator
 D Top plate of collecting chamber
 E Liquid air reservoir
 F Ion collector

FIGURE 4 SECTION OF COLLECTOR MOUNTING

maximum depth of 0.37 mm was made in tungsten by 20.4 mg of K^{39} ions, as shown in Fig. 5B. Thus it was necessary to replace the tungsten slit edges after each run. The sputtering of the target is less severe, for the beam is there spread over an area of 17×10 mm. For these 5000 volt ions the mass of the sputtered tungsten was approximately five times that of the impinging potassium. A coating of tungsten always forms on the collector along with the potassium, but it is not thick enough to introduce any complications.

This method of collecting has the disadvantage that the final isotopic sample is spread over an area of 5 cm^2 , but for many experiments this introduces no real difficulty. The inside of the collector can be lined with a thin sheet of nickel; this provides a simple way of removing the sample. If the sample is to be used for spectroscopic experiments its large area is not at all troublesome. If it is to be used for studies in nuclear physics a small target is required, but even with a large collector there is great enough surface density for such experiments. If it is desired to ascertain the radioactive properties of certain isotopes the large surface of the collector is no disadvantage at all, as will be shown in another section.

In all of the early experiments with this spectrometer the slit was placed normal to the central ray of the ion beam. Calculation shows that, in general, for this type of magnetic lens the focal plane makes an angle different from 90° with the central ray. For this particular lens the angle is calculated to be 54° . Accordingly



Sputtering of Slit Edges

Fig. 5

a mounting for the slit was constructed in which both edges of the slit were in the focal plane. Although S. S. W. located the focal plane, the resolution was not as good as was expected, so the author made another effort to locate this plane. A slit mounting was provided with brass rods which could slide through holes in brass blocks, as shown schematically in Fig. 6. By turning the shaft F (this could be done from outside the vacuum by means of the same conical greased vacuum-tight joint shown in Fig. 4) the plane A-A could be moved until it coincided with the focal plane. The intersection of the focal plane and the central ray of the ion beam proved to be 2.80 mm inside the end of the spectrometer case. This is 6 mm nearer to the pole pieces than was indicated by S. S. W.

The same collecting chamber was used in all of these experiments. It is shown, in simplified diagram, in Fig. 7, in its final form with the Geiger-Müller counter installed. The details of the collector finally used for potassium are shown in Fig. 10. The width of the focal plane slit is adjustable from outside of the vacuum by means of a lever mounted on a conical joint. No effort was made to show this mechanism in Fig. 7.

The Electrical Circuit

The electrical circuit of the spectrometer has been considerably improved. A vernier rheostat was installed in the field circuit of the high voltage generator to facilitate the adjustment of the accelerating potential. The focussing bars have been discarded, for

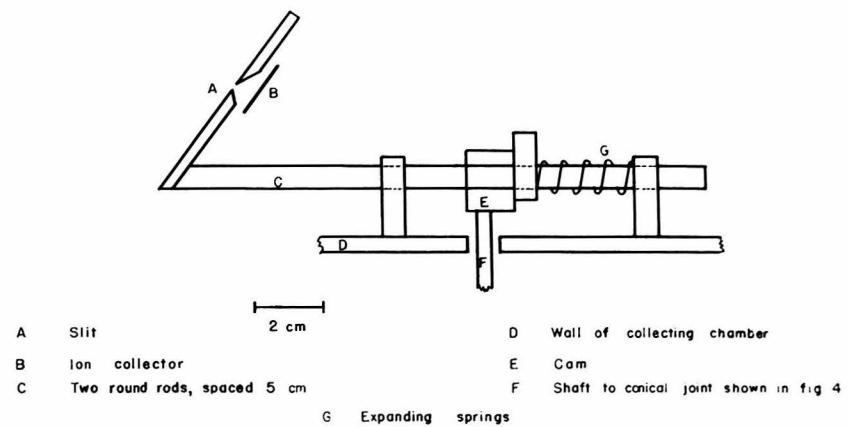


FIGURE 6 MOVABLE SLIT MOUNTING

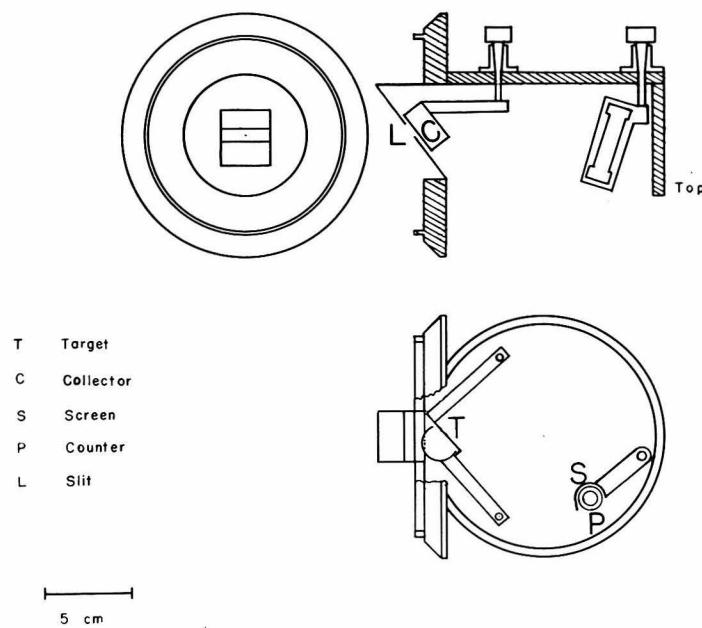


FIG. 7 COLLECTING CHAMBER
WITH COVER REMOVED

they proved to be unnecessary. The wiring diagram is shown in Fig. 8. R_1 is a 5000 ohm protective resistor, to limit the current in the event of a momentary gaseous discharge inside the apparatus. R_2 is a 60,000 ohm potential divider to adjust the potential of the focusing wire and to supply the proper voltage to the regulating circuit. V is a voltmeter and M a voltmeter multiplier.

The voltage from the potential divider is balanced against dry batteries and the difference is applied to the grid of an amplifying tube. This tube is direct-coupled to a second tube, the plate current of which controls a single pole double throw relay. For zero signal input to the amplifier the relay arm is in the middle position; if the signal increases the arm moves one way, and if it decreases the arm moves the other way. The relay controls a small direct current motor. The motor drives a winch which hoists two helices of chromel wire (0.5 cm diameter, number 18 B & S wire, altogether about 10 meters) in or out of vertical tubes of mercury. The helices are connected together at their tops; connections to the mercury columns are then terminals of a resistor which is controlled by the amplifier. This resistor is placed in the field circuit of the high voltage generator. By adjusting the battery voltage any desired sensitivity of the regulator can be attained. The generator voltage was constant within 0.2% with a 135 volt balancing battery, and this is all the sensitivity that was required.

A diagram of the circuit is shown in Fig. 8. The first stage

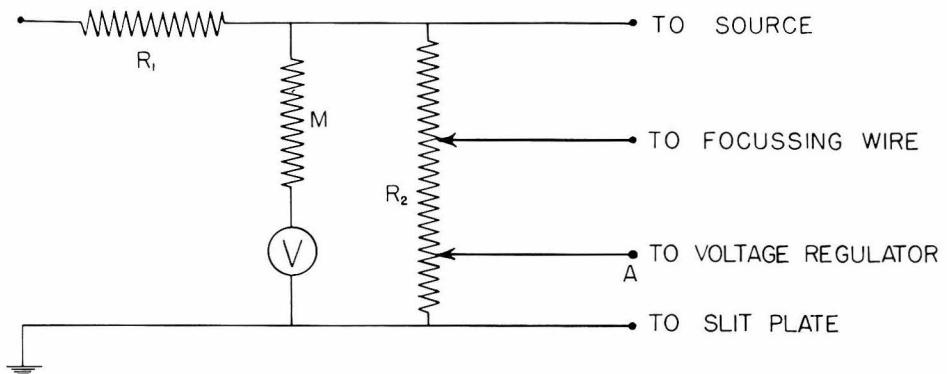


FIG. 8 WIRING DIAGRAM OF SPECTROMETER

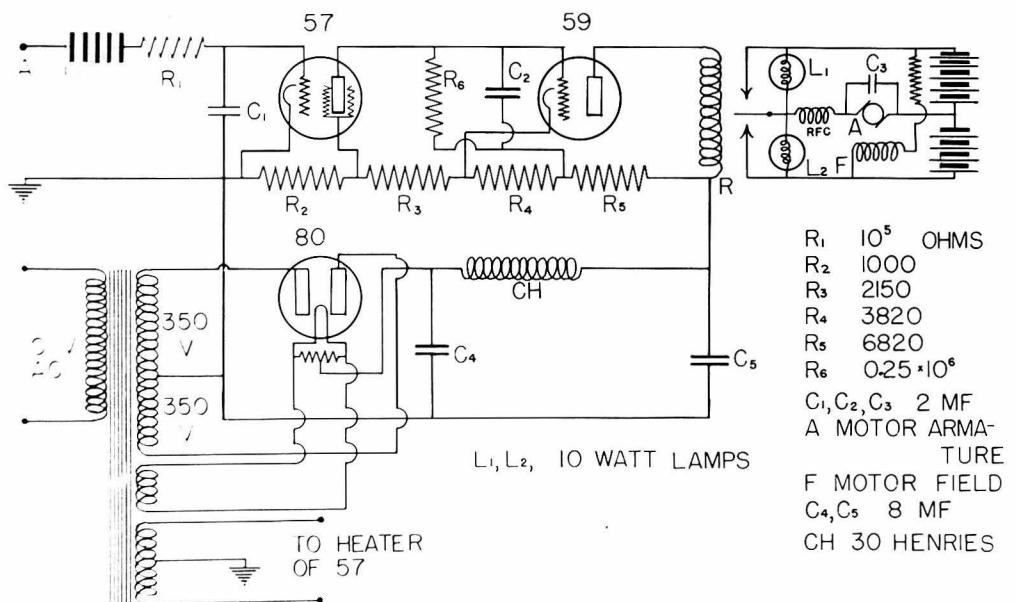


FIG. 9 VOLTAGE REGULATOR

of the amplifier is designed for high voltage amplification and the second stage is designed for high mutual conductance. The transconductance of the whole circuit is 260 milli-mhos. The condensers C_1 , C_2 shown prevent induced alternating currents from getting into the circuit and causing the relay to chatter. The lamps L_1 , L_2 across the relay contacts prevent sparking and also serve as convenient indicators to show what is going on. The radio frequency choke RFC and the condenser C_3 were necessary to prevent an amplifier (used with a Geiger-Müller counter) from transmitting a pulse to a mechanical counter whenever the relay moved.

Operation of the Instrument

When the source is first heated a large amount of gas is given off. Gas also comes off the walls of the apparatus wherever the ion beam strikes them. Fortunately the apparatus can be well outgassed by running the spectrometer at a very low intensity, that is, at a K^{39} current through a 4 mm slit at the focus, of about 1 microampere. This does not waste an appreciable amount of potassium and allows the isotope collection to be made at a better vacuum with the attendant better resolution.

For potassium the dispersion is 5.6 mm per mass unit. All potassium isotopes were collected using a 4 mm focal plane slit. The resolution is good at low intensities, but separation has never been obtained at high intensities. The largest collecting current (K^{39}) at which there is separation is 180 microamperes. In operation the

emission is always kept as high (by adjusting the heater current) as is consistent with good resolution. The limit of useful emission seems to be set by gas given off either by the magnesium oxide insulators around the heaters or by the emitter itself, for this gas produces scattering of the ion beam. A week of continuous collecting is required to collect a sample of a well resolved potassium isotope; in this time the yield is 20 mg of K^{39} , as indicated by the time-integral of the collecting current. Fig. 13 shows the resolution of the instrument at operating intensity (100 microamperes K^{39} current through a 4 mm slit).

The operation of the instrument is extremely stable. The spectrometer has run for ten hours without any attention whatever and without any appreciable change in either the collecting current or the mass arriving at the slit. The equation for focussing is

$$H^2 = 50.6 Vm$$

where H is the magnetic field in gauss, V the accelerating potential in volts, and m the atomic weight of the singly charged ions. The amount of iron in the core of the magnet is adjusted until the core is saturated; then it is a simple matter to keep the magnetic field constant, and for focussing we have the simple relation,

$$Vm = \text{constant.}$$

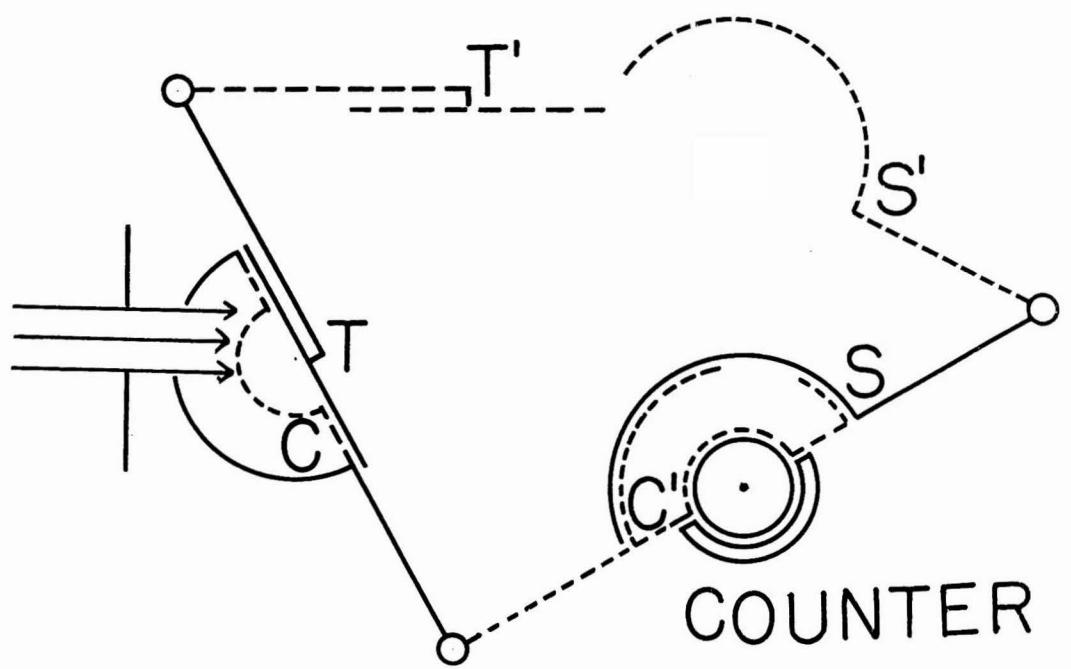


Diagram showing arrangement of collector,
target, and counter

Fig. 10

on the concave surface of the collector. The collector is partially closed at the ends. In Fig. 10 the collector, target, and screen S are shown in the collecting position, and the dotted lines indicate the counting position. In the collector-counting position the screen is moved so that it nearly covers the hole in the collector. This minimizes the error due to contamination of the walls of collecting chamber.

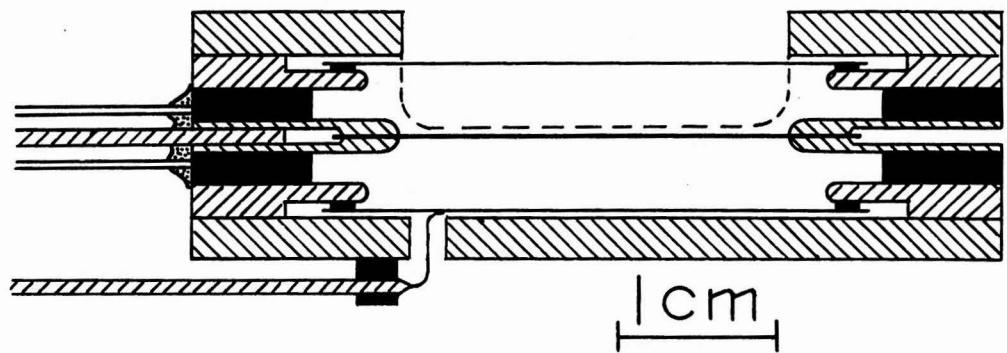
The Counter

There are two rather stringent requirements on the GM counter: it must have a sufficiently thin wall to pass most of the beta-particles emitted by potassium, and it must have an extremely low background. About thirty different counters were constructed, all of the same general type as the one finally adopted, but differing in details. All counters were built in such a way that the gas pressure inside and outside of the counter wall was necessarily the same, the plan being to admit air at the desired pressure to the whole spectrometer.

The early counters were constructed with a cylinder of 0.0127 mm aluminum foil. Some were also made of 0.0254 mm copper. The last counters were made of 0.0254 mm of aluminum. The copper cylinders were made by wrapping copper foil around a mandrel and soldering the overlapping parts together. The aluminum tubes were made in the same way except that they were held together by Duco cement, care being

taken not to get any cement inside the cylinder. The obvious way to construct such a counter seemed to be to mount the cylinder in a brass tube which was fitted with insulating plugs to support the central wire, enough of the cylinder being cut away to allow the beta-particles from the collector to reach the aluminum foil. Most of the counters tried were made in just this way, and none of these had a sufficiently low background. To ascertain whether the background was low enough, small known amounts of potassium salts were placed upon the collector and the number of beta-particles from these salts was measured. From these measurements one could estimate the number of beta-particles to be expected from the largest obtainable isotopic sample. Another significant shortcoming of these counters was their irregular and erratic behavior; for a long time this was very troublesome. Some experiments were then tried upon counters in which guard rings were used and in which there was no insulating material in strong electric fields. The counter shown in Fig. 11 was built and proved to be quite satisfactory; this is the one used in all measurements. The counting portion is 3 cm long and 1 cm in diameter. The background was 3 per minute, and remained remarkably steady.

Upon the suggestion of Dr. Bowen a five walled lead box, with walls 5 cm thick, was constructed and mounted in such a way that it surrounded the collecting chamber, except for one side, and the pole pieces of the spectrometer formed this side of the box. The lead weighed 360 kg and was mounted on rollers so that it could be easily



Detail of Counter

The insulating material, shown in black, is amber, the wire is steel, the cylinder is aluminum, and all other parts are brass.

Fig. 11

removed. With the lead around the counter the background was 1 per minute. It is known from measurements with an ionization chamber that two thirds of the penetrating radiation in this laboratory is gamma radiation, most of which would be stopped by the lead. Thus we have good evidence that the counter has the lowest possible background, and also that there are no spurious counts due to insulation leakage.

The absorption coefficients in aluminum of the beta-rays from potassium were found by Campbell and Wood¹⁹⁾ to be 75 and 29 cm^{-1} for the two bands. This means that the transmissions of the counter wall are 83% and 93% for the two bands.

There was of course no difficulty about adjusting the gas pressure in the system preparatory to the count that preceded collection, but during the operation of the spectrometer a film of potassium is formed on most of the surfaces in the interior of the apparatus, and it was clear that if air should be admitted to the apparatus its composition would change continuously until the potassium was completely oxidized, or until the oxygen was gone. For this reason it seemed advisable to use a different gas in the counter. Other gases tried were hydrogen, helium, nitrogen, and mixtures of these. None of these were at all satisfactory: the counter evidently required oxygen. With any gas other than air in the system there was no large enough range of voltage for which the counter background was independent of voltage. Thus it was necessary to oxidize the potassium before

any counts could be taken. This was done by admitting fresh dry air to the spectrometer at intervals of 5 hours until equilibrium was reached.

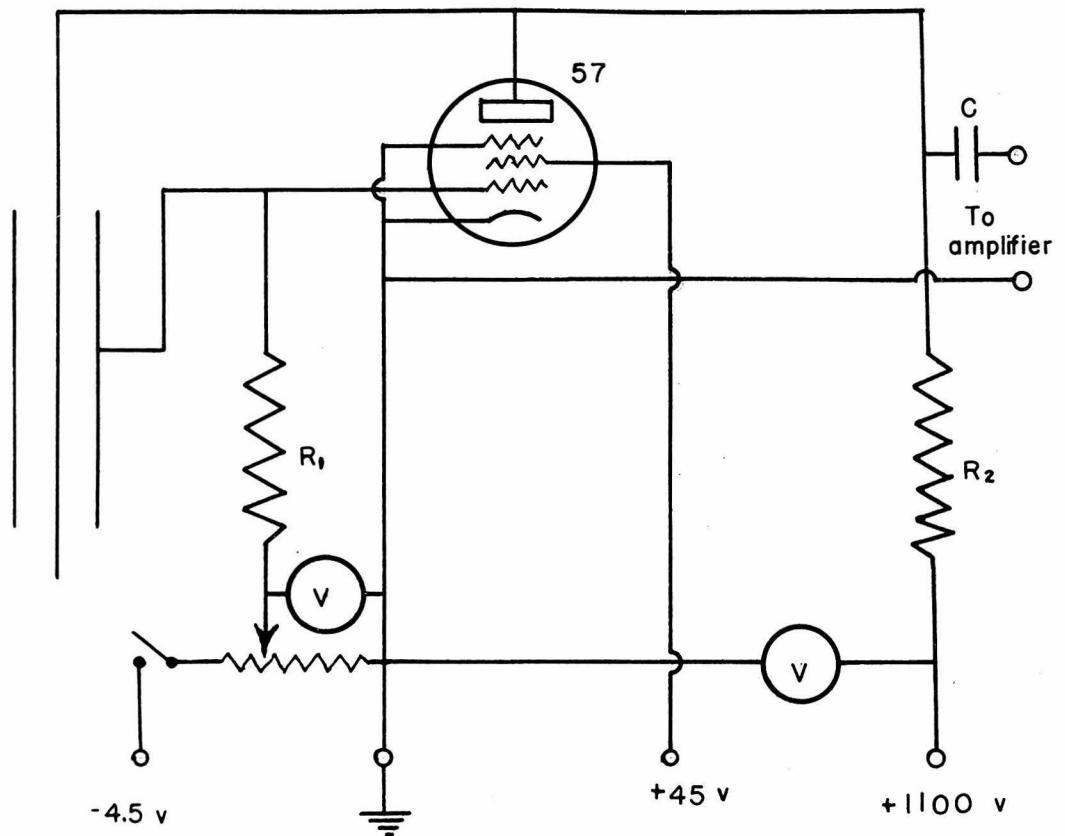
With 1100 volts applied to the counter (with the wire positive) the counting range was 100 volts wide. The voltage was carefully regulated by means of the regulator which has already been described.

The Counter Circuit

The circuit devised by Neher and Harper¹⁰) for coupling the counter to the amplifier was of considerable value in this experiment, for with this circuit no special precaution need be taken with the insulation of the counter. The counter was coupled to an audio-frequency amplifier, the output of which operated a telegraph relay, which in turn operated an impulse counter. A diagram of the coupling circuit is given in Fig. 12.

Measurements

The actual measurement of the activity of the deposit of the collector must be made in such a way that it is independent of small changes in voltage, pressure, and temperature of the counter. This was done by measuring the counting rate first with the collector near the counter, then with the screen near the counter, and taking the ratio of these rates; the same ratio is measured before and after deposition of potassium. Actually there may be small slow changes



R_1 20 megohms

R_2 2 megohms

C 50 cm

FIG. 12 COUNTER CIRCUIT

in the counter sensitivity during the counts, so the counts with the collector and screen near the counter were taken in alternate half hour intervals, usually over a period of 14 hours.

For a given set of counts let N_c be the total number of collector-counts and N_s the total number of screen counts. The counting ratio is $R = N_c/N_s$. It is shown by Dr. Bateman²¹⁾ that if the average counting rate for a given interval is x , the probability that n counts will occur is

$$P_n = x^n e^{-x} / n!,$$

and that the square root of the most probable value of the square of the deviation from this value is x . Now this probability P_n is not the Gaussian distribution, but for a total number of counts $N > 100$ it differs very little from the Gaussian distribution. This condition is satisfied in all of the data to be presented here, for $N_c > N_s > 400$, so that we can calculate the probable error as if the distribution were Gaussian. Thus the probable error in N counts is $0.675 \sqrt{N}$ and the probable error in R is $0.675 R(1/N_c + 1/N_s)^{1/2}$. It is this probable error that will be indicated along with counting ratios.

Two complete sets of measurements were made on potassium. The first set was completed without opening the apparatus, with each sample of potassium deposited on top of the previous one, so that the count was cumulative. The results are shown in Table I.

Table I*

Collector deposit	Counting ratio	Probable error
No deposit	1.04	0.05
(1) 4.35 c/14 mg K ⁴¹	1.04	0.05
(2) 4.06 c/8300 mg K ⁴⁰ + (1)	1.28	0.06
(3) 4.7 c/l mg K ³⁹ + (1) + (2)	1.30	0.06

In the second set of measurements larger deposits of each isotope were made. This involved opening and cleaning the apparatus and renewing the source between runs. The K³⁹ sample of Table II was known to be contaminated with more than the usual amount of K⁴⁰.

Table II

Collector deposit	Counting ratio	Probable error
No deposit	1.07	0.05
13.4 c/14 mg K ⁴¹	1.14	0.06
No deposit	1.10	0.05
20.4 c/8300 mg K ⁴⁰	1.93	0.08
No deposit	1.03	0.05
13.6 c/l mg K ³⁹	1.26	0.06

If all the collector current were due to K⁺ ions and if all these ions, after neutralization, were deposited on the collector,

* The abundance ratios of the potassium isotopes were measured by A. O. Nier and by A. K. Brewer.^{9,10}

the factor c in Tables I and II would be one. Somewhat indirect measurements indicate that from 5% to 15% of the collector current is due to escape of secondary electrons. Visual inspection of the deposit on the collector and target indicates that from 60% to 80% of the potassium striking the target adheres to the collector. Thus the factor c appears to lie between 0.5 and 0.75.

The data of Table III were obtained from two counts made on a nickel sheet when it was coated with a thin layer of KCl and the second after this had been washed off with distilled water.

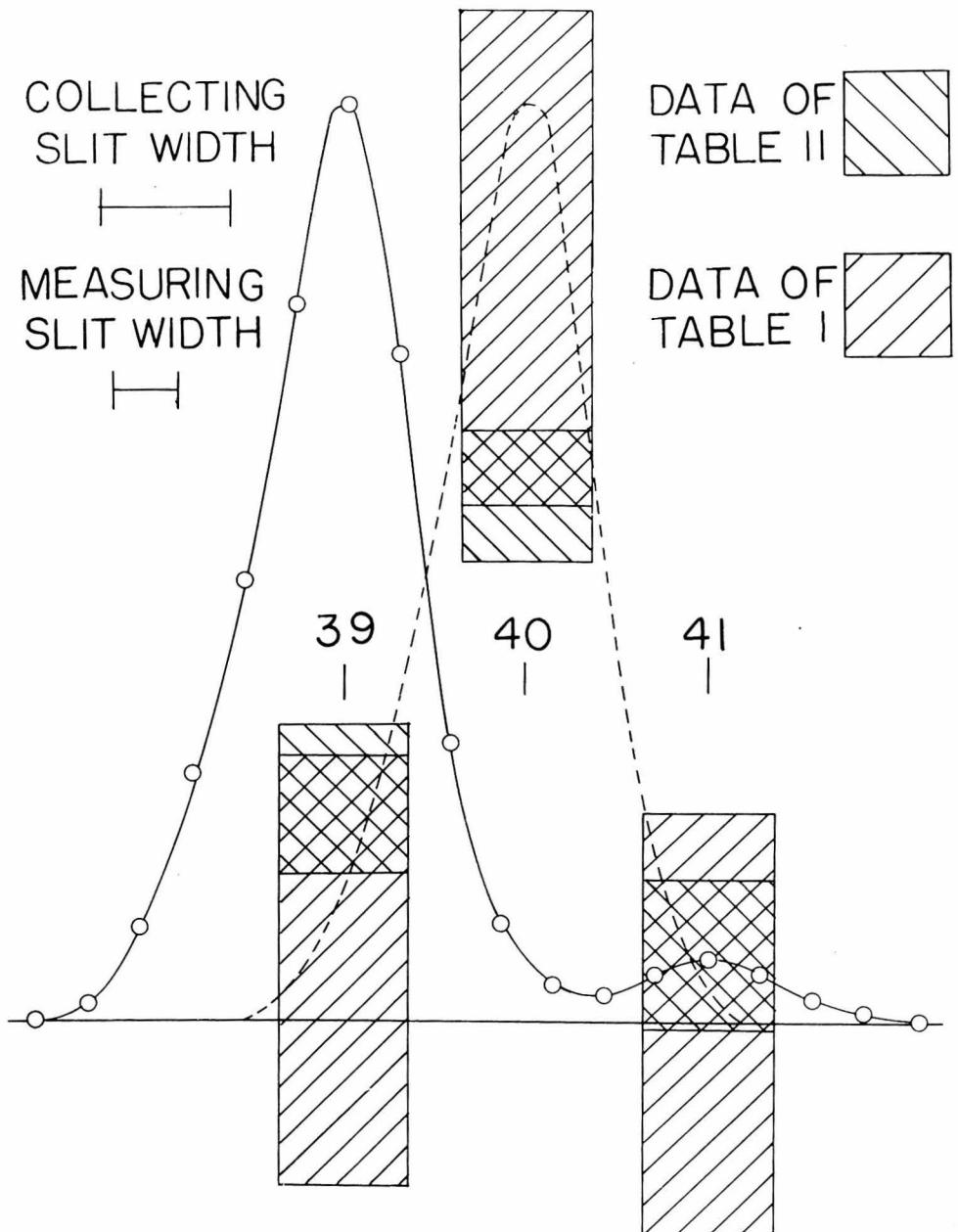
Table III

Collector deposit	Counting ratio	Probable error
67.2 mg KCl on Ni foil	3.22	0.12
Clean Ni foil	1.03	0.05

Calculations and Results

The activity of each sample is defined as $R - R_0$, where R_0 and R are the counting ratios taken before and after deposition on the collector. The significance of $R - R_0$ can be seen from the following:

Let N = the background of the counter,
 N_1 = the count due to the screen,
 N_2 = the count due to the collector,
and N_3 = the count due to the deposit.



Resolution Curve and Counting Data

Fig. 13

Then $R_o = (N + N_2)/(N + N_1)$, $R = (N + N_2 + N_3)/(N + N_1)$, and these ratios are independent of the voltage of the counter. The difference $R - R_o = N_3/(N + N_1)$ is thus proportional to the number of beta-particles emitted by the deposit.

The data of Tables I and II are presented graphically in Fig.

4. The solid curve, giving the collecting current plotted against mass, is measured with a 2 mm slit and shows the resolution maintained during the collection of all samples except the K^{39} sample of Table II. The width of the cross hatched areas shows the positions of the collecting slit. The ordinates of the centers of the cross hatched areas are the measured activities, and the lengths are twice the probable errors, all reduced to samples of the same size. All scales of ordinates are arbitrary.

There are 32.9 mg of K^{39} in 67.2 mg of KCl. The activity of this sample, after correcting for absorption of both bands of beta-particles in KCl on the assumption that the mass absorption coefficients are the same as for aluminum,¹⁹⁾ is 2.58. The correction for absorption in tungsten in Tables I and II is quite negligible. Measurements by Bocciarelli²²⁾ show that 60% of the emitted beta-particles lie in the low energy band. Taking the weighted mean of the measured activity of K^{40} from Tables I and II, reducing to a standard sample of 20 mg, and comparing with the measurements of Table III, also reduced to 20 mg, we have, for the three possible assumptions as to which bands belong to K^{40} , the results:

Hypothesis	Measured activity $c = 0.75$	Expected activity $c = 0.5$
K^{40} emits high energy band	1.08	1.62
K^{40} emits low energy band	1.08	1.62
K^{40} emits both bands	1.08	1.62

This clearly indicates that K^{40} is responsible for the entire known activity of potassium.

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