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# The Radioactive Isotope of Potassium

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By means of a high intensity mass spectrometer we have separated the three isotopes of potassium. Measurements of the radioactivity of the isotopic samples indicate that $K^{40}$ is radioactive. Comparison of the beta-particle emission from the isotopic samples and from KCl shows that $K^{40}$ is responsible for both of the known beta-ray bands.

## INTRODUCTION

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. Dobrosserdow[2] 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 Thomson[3] in 1921 that potassium has two isotopes, $K^{39}$ and $K^{41}$, made it of interest to ascertain which of these is radioactive. G. v. Hevesy and M. Logstrup[4] 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, $Ca^{39}$ or $Ca^{41}$, of this disintegration, were unsuccessful. This suggested the possibility that a rare isotope $K^{40}$ is responsible for the radioactivity, with the well-known $Ca^{40}$ as the end product. Further measurements with a Geiger-Müller counter by G. v. Hevesy, W. Seith, and M. Pahl[5] on the heavy samples of potassium showed an increase in the radioactivity of 4.43±0.53 percent. A determination of the atomic weight by Hönigschmid and Goubeau[6] indicated an increase in the concentration of $K^{41}$ of 4.2±0.84 percent. They asserted that these results prove the radioactivity of potassium to be due to $K^{41}$ and that they definitely exclude a hypothetical $K^{40}$. More precise measurements of the atomic weight of potassium[7] and of v. Hevesy's heavy potassium,[8] together with recent measurements of the abundance ratio[9, 10] of $K^{39}$ and $K^{41}$, raised doubts as to the correctness of v. Hevesy's conclusion:* they showed that the fractional increase in concentration of $K^{41}$ was 10.6 percent. 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 $K^{40}$. Klemperer[11] and Newman and Walke[12] considered that of the hypothetical group, $K^{39}$, $K^{40}$, $K^{41}$, $K^{42}$, the only one which could give the observed activity was $K^{40}$, and that this is consistent with v. Hevesy's observations if the latest values of atomic weights and abundance ratios are used. After the discovery of $K^{40}$ Sitte[13] pointed out that, since $Ca^{40}$ and $A^{40}$ 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

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[1] J. J. Thomson, Phil. Mag. (6) **10**, 584 (1905).
[2] D. Dobrosserdow, Phys. Ber. **7**, 1414 (1926).
[3] F. W. Aston, Phil. Mag. **42**, 436 (1921).
[4] G. v. Hevesy and M. Logstrup, Zeits. f. anorg. allgem. Chemie **171**, 1 (1928).
[5] G. v. Hevesy, W. Seith and M. Pahl, Zeits. f. physik. Chemie, Bodenstein Festband, 309 (1931).
[6] O. Hönigschmid and Goubeau, Zeits. f. anorg. allgem. Chemie **163**, 93 (1927); **177**, 102 (1928).
[7] G. P. Baxter and W. M. MacNevin, J. Am. Chem. Soc. **55**, 3185 (1933).
[8] G. P. Baxter and C. M. Alter, J. Am. Chem. Soc. **55**, 3270 (1933).
[9] A. O. Nier, Phys. Rev. **48**, 283 (1935).
[10] A. K. Brewer, Phys. Rev. **48**, 640 (1935).
* G. v. Hevesy in a recent article predicts a radioactive isotope, $K^{40}$. Naturwiss. **23**, 583 (1935).
[11] O. Klemperer, Proc. Roy. Soc. **A148**, 638 (1935).
[12] F. H. Newman and H. J. Walke, Phil. Mag. **19**, 767 (1935).
[13] K. Sitte, Zeits. f. Physik **96**, 593 (1935).

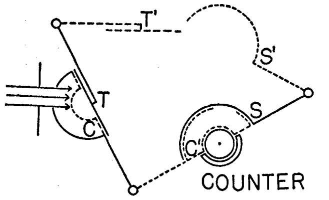

FIG. 1. Solid lines show collector, $C$, in collecting position and screen, $S$, in counting position for $R_0$. $C'$ and $S$ show collector and screen in counting position for $R$.

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.

## APPARATUS

Of these instruments only the high intensity mass spectrometer described by Smythe, Rumbaugh, and West[14] gives sufficient intensity. Minor changes have been made in the instrument. The heating elements now consist of tungsten helices enclosed in MgO tubes,[15] constructed by West. The Kunsman $K^+$ ion source is now prepared by fusing the nitrates of potassium and iron together and igniting. The resultant oxide mixture is reduced in hydrogen, pounded into the grooves in the source block until it forms a layer of solid metal, placed in the spectrometer and is again reduced in hydrogen. After the spectrometer is evacuated the source is outgassed for a day before using. An automatic voltage regulator, which controls the field current of the generator supplying the accelerating potential for the ions, keeps the voltage, usually about 5000, constant within 0.2 percent. The only attention required by the spectrometer after it is set for collecting a given mass is the adjustment every eight hours of the source heater current and the magnetic field current, and the renewal of the liquid air on the mercury trap.

The collecting end of the apparatus has been entirely changed. $K^+$ ions have a very destructive effect on any target upon which they impinge, so that not only the potassium but also the material of the target is sputtered on all neighboring objects. To collect the potassium we set a tungsten target at an angle such that few of the sputtered atoms would retrace the path along which they came as ions, and surrounded it by a box through a hole in which the positive rays entered, as shown in Fig. 1.

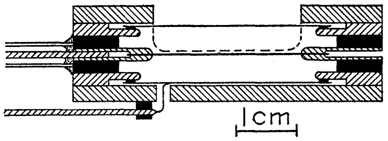

FIG. 2. Longitudinal section of Geiger counter showing amber insulation in black and brass parts cross hatched. The dotted line outlines the exposed area of aluminum foil wall of the counting region. A cross section of the counter appears in Fig. 1.

A Geiger-Müller counter seemed to be the most satisfactory instrument for measuring the emitted beta-particles. Four requirements are imposed upon this counter. First, to minimize its background its volume must be small. Second, to avoid absorption of beta-particles the walls must be thin. Third, the counter must subtend a large solid angle at the collector. Fourth, to avoid possible contamination the collector should be handled as little as possible, preferably not at all. After a few unsuccessful experiments with external counters we followed Mr. Harper's suggestion that we make a counter which is not vacuum-tight, and place it inside the collecting chamber, admitting air to the whole apparatus to adjust the pressure in the counter.

The design of the thin-walled counter is shown in Fig. 2. The cylinder, 3.5 cm long, was made by wrapping 0.0254 mm aluminum foil around a mandrel 0.93 cm in diameter and fastening the overlapping portions with Duco cement. The central wire is steel, 0.254 mm in diameter. The cylinder is mounted in a brass tube which is cut away on one side to expose the aluminum. The mounting is designed to shield electrostatically the sharp ends of the aluminum and to guard it against leakage from the wire across the amber insulators. The background of all counters not so constructed

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[14] W. R. Smythe, L. H. Rumbaugh and S. S. West, Phys. Rev. **45**, 724 (1934).

[15] W. H. Swanger and F. R. Caldwell, Nat. Bur. Stand. J. Research **6**, 1131 (1931).

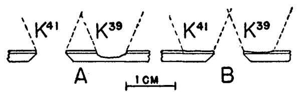

FIG. 3. *A* shows the destructive action of 13.5 mg of 5000 volt K$^{39}$ ions striking a brass slit faced with 0.15 mm of molybdenum. The maximum penetration from the surface is 1 mm. *B* shows the action of 20.4 mg of the same ions striking a brass slit faced with 0.5 mm of tungsten. The maximum penetration from the surface is 0.37 mm.

was large and unsteady, but that of the counter used is extremely steady. A lead box with walls 5 cm thick and open at one end was constructed to slide over the whole collecting chamber. The massive iron pole-pieces of the spectrometer formed the sixth side of the box. The background of the counter was 1 count per minute when surrounded by the lead and 3 counts per minute when not. This is just the ratio of cosmic-ray ionization to total ionization in this laboratory and indicates that the counter background is an absolute minimum.

The circuit devised by Neher and Harper[16] for coupling the counter to the amplifier proved to be of great value, for it requires no especially good insulation of the counter.

The transfer of the collected sample from the collecting position to the counting position is accomplished by the arrangement shown in Fig. 1. The collector *C*, covering the face of the target *T*, is a half-cylindrical box, partially closed at the ends, with a hole through which the positive rays can enter. Except for the small amount escaping through this hole and the ends, and adhering to the target outside of the bombarded area, all the potassium impinging on *T* is collected on the inside of the box. During the collection the counter is protected by a cylindrical screen, *S*. The screen, target, and collector may be turned, in this order, to the position shown by the dotted lines. The screen may then be turned back to cover the hole in the collector. Thus all measurements are independent of the contamination of the walls of the collecting chamber. The procedure for investigating a single isotope is as follows: after the apparatus has been evacuated long enough to remove the moisture, air is admitted to 5.6 cm pressure and counts are made alternately on *S* and *C* for one-half hour intervals until at least 400 counts have been made on each; the apparatus is then evacuated and the sample collected on *C*; air is admitted and pumped out several times until the potassium, which always collects on the walls of the apparatus near the source, is so oxidized that it no longer removes oxygen from the air rapidly (the counter would never operate stably without oxygen); alternate counts are then made on the collector and screen as before. During a set of counts the counter is operated at a fixed voltage and pressure. The collected sample is always mixed with tungsten sputtered from the target. There seems to be no easy way of eliminating this sputtering without adopting an entirely different collecting technique.

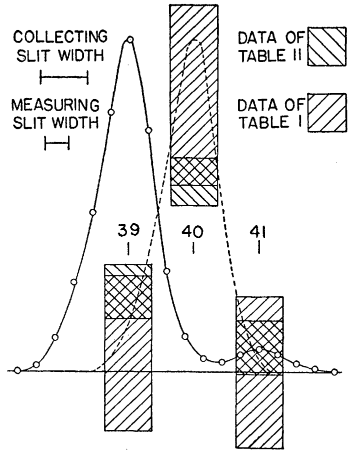

FIG. 4. The solid curve, giving the collecting current plotted against mass, is measured with a 2 mm slit and shows the resolution maintained during the collection of all samples except the K$^{39}$ sample of Table II. The width of the cross-hatched areas shows the positions of the collecting slit. The ordinates of the center 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.

When collecting, a slit which is set in the focal plane of the spectrometer 3 mm in front of the hole in the collector is opened to 4 mm. The destructive effect of the intense positive ion beam on the slit material is shown in Fig. 3. The slit edges were originally of 1.6 mm beveled brass. The ion beam widened a 4 mm slit to 5.5 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. Tungsten proved to be more resistant under ion bombardment: a hole 4×6 mm in

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[16] H. V. Neher and W. W. Harper, Phys. Rev. **49**, 940 (1936).

area and with a maximum depth of 0.37 mm was made in tungsten by 20.4 mg of $K^{39}$ ions. Thus it was necessary to replace the tungsten slit edges after each run. The sputtering of the target was 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.

All collections were made at good resolving power, except the last item in Table II. The curve of positive ion current plotted against atomic weight, taken with a focal plane slit 2 mm wide, is shown in Fig. 4. The usual collecting current, with a 4 mm slit, is about 0.12 ma. Although greater collecting currents are easily obtained by increasing the source temperature, this usually broadens the peak and greatly shortens the life of the source. The maximum amount of potassium that can be collected in one run is not known: at the end of the run, one week long, in which the 20.4 mg sample was collected the collecting current was still near its maximum, the resolving power was good, and there was no sign of exhaustion of the source.

## MEASUREMENTS

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. The *counting ratio* $R$ in each case is the ratio of collector-counts to screen-counts. The error is computed in the standard way:[17] the probable error in $N$ counts is $0.675\sqrt{N}$; the probable error in $R$ $(=N_2/N_1)$ is $0.675R(1/N_1+1/N_2)^{\frac{1}{2}}$.

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 results are shown in Table II. Unfortunately the resolving power went bad several times during the collection of the $K^{39}$ sample, perhaps because a different batch of emitter and new heater insulation were used, so that it is contaminated with an unknown amount of $K^{40}$.

TABLE I. *β-particle counts from the collector as deposits of $K^{41}$, $K^{40}$ and $K^{39}$ were added successively.*

| 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 $c/1$ mg $K^{39}+(1)+(2)$ | 1.30 | 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 factor $c$ in Tables I and II would be one. Somewhat indirect measurements indicate that from 5 percent to 15 percent of the collector current is due to the escape of secondary electrons. A visual inspection of the deposit on the collector and target indicates that from 60 percent to 80 percent 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.

## 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.

Then $R_0=(N+N_2)/(N+N_1)$, $R=(N+N_2+N_3)/(N+N_1)$, and these ratios are independent of the voltage of the counter. The difference,

TABLE II. *β-ray counts from separate deposits of $K^{41}$, $K^{40}$ and $K^{39}$.*

| 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 $c/1$ mg $K^{39}$ | 1.26 | 0.06 |

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[17] H. Bateman, Phil. Mag. **20**, 704 (1910).

TABLE III. *β-ray counts from natural potassium.*

| COLLECTOR DEPOSIT | COUNTING RATIO | PROBABLE ERROR |
|---|---|---|
| 67.2 mg KCl on Ni foil | 3.22 | 0.12 |
| Clean Ni foil | 1.03 | 0.05 |

$R-R_0=N_3/(N+N_1)$, 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.

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 the beta-particles in KCl on the assumption that the mass absorption coefficients are the same as for aluminum,[18] is 2.58. The correction for absorption in tungsten in Tables I and II is quite negligible. Measurements by Bocciarelli[19] show that 60 percent of the emitted beta-rays lie in the low energy band and the remainder in the high 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 activity to be expected from 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 shown in Table IV. This clearly indicates that $K^{40}$ is responsible for the entire known activity of potassium.

TABLE IV. *Comparison of the measured activity of $K^{40}$ with that to be expected upon various hypotheses.*

| HYPOTHESIS | MEASURED ACTIVITY | | EXPECTED ACTIVITY |
|---|---|---|---|
| | $c=0.75$ | $c=0.5$ | |
| $K^{40}$ emits high energy band | 1.08 | 1.62 | 0.63 |
| $K^{40}$ emits low energy band | 1.08 | 1.62 | 0.94 |
| $K^{40}$ emits both bands | 1.08 | 1.62 | 1.57 |

### ACKNOWLEDGMENTS

We wish to acknowledge our debt to those who have worked on this problem in its earlier stages, particularly Dr. West, and to Mr. Harper, who lent us his amplifier and assisted in some of the counter work. We are grateful also for the assistance in the laboratory of Mr. R. H. Mansfield.

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[18] N. R. Campbell and A. Wood, Proc. Camb. Phil. Soc. **14**, 15 (1907).

[19] D. Bocciarelli, R. C. Accad. Lincei (6) **17**, 830 (1933).

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# The Disintegration of Beryllium by Protons*

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The disintegration of Be by protons of energies from 45 to 125 kv has been studied. The yield curve for a thick target has been determined and the ratio of the number of alpha-particles and deuterons ejected from the target has been determined for various voltages. The ratio was very close to unity. The ranges of both groups of particles were found to be 7.1 mm. From the energy relations the mass of $Be^8$ was found to be 8.0074. The experimental value of the effective area for collision was found to be of the same magnitude as that predicted by Gamow's theory of the penetration of a potential barrier by protons having zero angular momentum.

ALTHOUGH much work has been done on the disintegration of Be by protons, it seemed worth while to investigate the two possible reactions in order to determine their relative probability of occurrence. According to Oliphant, Rutherford and Kempton,[1] the two reactions are:

$$ {}_4Be^9+{}_1H^1={}_3Li^6+{}_2He^4, \tag{1}$$

$$ {}_4Be^9+{}_1H^1={}_4Be^8+{}_1H^2. \tag{2}$$

They found that the $H^2$ and the $He^4$ particles

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* A dissertation submitted to the faculty of the division of the physical sciences in candidacy for the degree of Doctor of Philosophy. Private edition, distributed by the University of Chicago Libraries, Chicago, Illinois.

[1] Oliphant, Rutherford and Kempton, Proc. Roy. Soc. **A150**, 241 (1935).