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Artificial Radioactivity Produced by Alpha-Particles

LOUIS N. RIDENOUR AND W. J. HENDERSON*
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
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Half-lives and thick target yields have been measured for some of the radioactive product reactions accessible with 9 Mev alpha-particles accelerated in a cyclotron. The radioactive isotopes are N^{13}, F^{17}, Al^{28}, P^{30}, Cl^{34}, K^{38}, Fe^{53}(?), Mn^{56}, Cu^{61}, Cu^{62}, Zn^{63}, Ga^{66}, Ga^{68}, Br^{78}, Rb^{82}, Rb^{84}. The complete data are given in Table II.

1. INTRODUCTION

THIS paper is a report of a preliminary survey of the radioactivity produced by alpha-particles in some of the lighter elements. The alpha-particles employed in the activation were accelerated in a cyclotron, and the available bombardment time was in consequence limited to a few hours. No attempt was made to seek short-lived radioelements, even in cases where they are known to be formed (e.g., sodium). It may be said, then, that no radioelement of half-life less than one minute nor more than one day would have been found in the present investigation, unless the reaction producing it were exceptionally probable. In point of fact, no radioelement of half-life outside these limits was found. In several cases (see Section 6), weak activities were found but have not yet been investigated further.

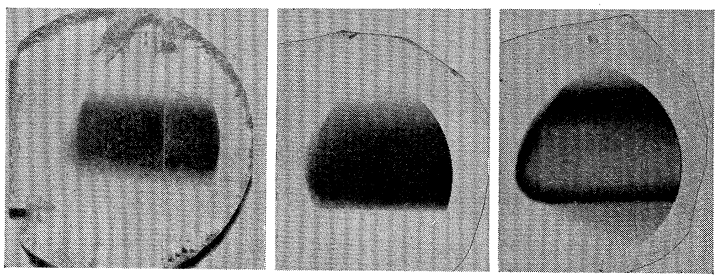
In all cases where a strong activity was found, an effort has been made to measure the half-life as well as the available intensity will allow, to assign the activity to the proper isotope by comparison with existing data and by chemical experiments, to measure the thick target yield of the reaction producing the radioelement, and, in three cases, to obtain an estimate of the energy of the emitted beta-particles by a measurement of their absorption in aluminium. The sign of the beta-particles has also been determined in most instances, by reversal of a magnetic field situated between the active source and a thin walled Geiger-Mueller counter.

2. THE ALPHA-PARTICLE BEAM PRODUCED BY THE CYCLOTRON

The distribution in energy of the alpha-particles emerging from the cyclotron used in the present work has been measured by the interposition of mica absorbing foils in the path of the beam. The measurement is described in an accompanying paper.[1] The total inhomogeneity of the beam amounts to about 1 Mev, the mean range of the particles corresponding to an energy of 9.0 Mev. In all the work reported here, save for a few of the earlier half-life measurements, the full energy of the beam has been employed; targets have been bombarded in a chamber which communicates directly with the main

* Now at Purdue University, West Lafayette, Indiana.

[1] M. C. Henderson, and White, Phys. Rev. To be published soon.



a b c

FIG. 1. Position of beam on target. *a*. Rocksalt sample bombarded for about one minute. Blackened area shows the location of the greater part of the beam. *b*. Record obtained on a piece of film cut roughly to size and shape of target, wrapped in 0.001 inch Al foil, placed in target holder and exposed to alpha-particle beam for about 1/20 second. Sensitivity is greater here than in case of rocksalt; note that traces of beam appear above the rectangular area blackened on the salt. *c*. Film exposed to beam in the same fashion as *b*; exposure time about 2 minutes. Traces of beam now appear below main spot. Note reversal in center, where beam is strongest.

vacuum tank of the cyclotron through a one-inch brass gate valve. The gate valve has been made vacuum tight by the replacement of its packing with sylphon bellows, and when the space between the gates is exhausted with a Hyvac pump, the pressure in the cyclotron vacuum tank can be maintained at 2×10^{-6} mm Hg while the target chamber is opened to the air for the insertion or removal of a target.

The beam currents measured by a Faraday cage inside the gate valve are of the order of 0.04 microamperes; however, the beam is considerably wider than one inch in a horizontal direction, so that the currents measured by using the target itself as a Faraday collector ranged between 0.01 and 0.02 microamperes, the rest of the ion beam having been intercepted by the gate valve. At the target, the bean measures about 1.2 cm vertically by 2 cm horizontally. The standard targets used in the yield measurements were round, and 3.7 cm in diameter. The position of the active area on the target is shown in Fig. 1.

It is reasonable to inquire whether the measured beam current may not be partly due to secondary electron emission from the target, since the target was simply a flat surface, without any shields to trap secondary electrons. It seems unlikely that secondary emission would play an important role, since the maximum energy which may be imparted to an electron by an alpha-particle of energy 8.8 Mev is 5100 volts. The weakest magnetic field anywhere on the surface of the target was 3500 gauss, in which the radius of curvature of a 5100-volt electron is 0.45 mm. Tests were conducted in which the yield of the reaction $Co^{59}(\alpha, n)$ Cu^{62} was measured (*a*) for a Co target in the position shown in Fig. 1 relative to the beam, and (*b*) for a Co target moved away from the gate valve far enough so that the beam overlapped the inner edge of the target by about 2 mm, due to the curvature of the alpha-particle beam in the stray field of the cyclotron magnet. If secondary emission is important at any time, its influence should be much greater in case (*b*) than in (*a*), resulting in the measured yield of the reaction being smaller in (*b*) than in (*a*). It was found that the yield in case (*b*) was only 6.5 percent smaller than that in case (*a*), so that one is presumably justified in assuming that the errors introduced by taking the measured beam currents to represent true ion currents to the target are much smaller than those arising from other causes (cf. Section 4).

3. HALF-LIFE AND ABSORPTION MEASUREMENTS

In all cases except that of Cr bombarded with alpha-particles, the half-lives given in this paper have been measured with a Lauritsen type electroscope provided with a window of aluminium 0.001 inch thick. Indeed, measurability with the electroscope has been the criterion serving to differentiate those activities selected for study from the others which are briefly noted in Section 6. The window of the electroscope is 3.8 cm square, while the active area on the targets, as already noted, is 1.2 cm$\times$2 cm (Fig. 1). The electroscope has been used window uppermost for the half-life and yield measurements, and the

target laid, active side down, in a standard position on the window.

For the beta-ray absorption measurements, the electroscope was used with the window down; the target, active side up, was placed so that its active area was in the middle of the window. The distance from target to window was about 1 cm. The square Al absorbers used were 5.3 cm on a side, and were simply laid centrally over the target.

4. Measurement of the Thick Target Yields

In the case of some of the stronger activities, the yield of the reaction producing the radioelement has been measured. To be accurate, such a measurement demands a knowledge not only of the total number of alpha-particles which have struck the target, but also of the time-distribution of the beam current. If the activation of samples be conducted with a naturally radioactive alpha-particle source, the number of particles striking the target each second decreases with the decay of the source in a known manner, and the correction for this effect can be easily computed from one measurement of the intensity of the source and a knowledge of its half-life. In our experiments, on the other hand, the fluctuations (as much as half-full beam) in ion current due to the "tuning" of the cyclotron were somewhat erratic and not negligible. Since particles which strike the target late in the bombardment have more influence on the radioactivity measured at the close of the irradiation than do those striking the target at the beginning, it is plain that some means of registering the beam current as a function of time is demanded.

A reflecting galvanometer was accordingly arranged to record the beam current on positive motion picture film. Timing marks about 10 inches apart were put on the film each minute, and at the close of every other run a standard deflection, produced by a measured voltage through a known resistance, was recorded on the film. After the film had been developed the area between the zero line and the galvanometer trace was cut out with shears, cut into lengths short compared with the half-life of the radioelement concerned, and the weight of each piece determined. The calibration records were also cut out and weighed, so that each gram of beam record corresponded to a known number of microcoulombs of doubly charged He ions striking the target. The charge to which each piece of the film record corresponded was multiplied by a fraction which took account of the decay occurring in the activity of the target during the interval between the average time represented by that particular part of the film record and the "zero time" at which bombardment was stopped. The sum of the corrected weights of the pieces of the film record then provides a measure of the effective number of alpha-particles which struck the target; the observed radioactivity would have been produced by the effective number of alpha-particles striking the target all at once at zero time.

While this procedure is somewhat complicated, it is believed that no errors comparable with those introduced by other causes have resulted from it. The five calibrations which were made, for example, agreed among themselves to better than 5 percent, demonstrating that the accuracy with which the films were cut out was good enough for the present purpose. The chief uncertainty in the relative values of the yields for different elements probably arises from our lack of knowledge as to the reflecting power of the body of any given target for electrons which are emitted from the thin active layer on its surface in such directions that they would not enter the electroscope without having been reflected. This uncertainty, of course, enters also into the determinations of absolute yields, where the additional uncertainty of the calibration of the electroscope in beta-particles per division exists.

The activity of the various targets bombarded for yield determination has always been referred to zero time—the time at which bombardment was stopped—by graphical projection back to zero time of the measured decay curve. Since the measurements of the activity of a target commenced not later than 2 minutes after the cessation of bombardment, any errors introduced by this extrapolation are not regarded as serious.

The calibration of the electroscope in beta-particles per division was carried out with a source consisting of 124 mg of powdered uranyl nitrate arranged over an area of the size and

shape of the active area on the targets (Fig. 1, *a*), on a piece of 0.002 inch Al foil cut out to the standard target size. The beta-particles entering the electroscope must have penetrated a minimum of 0.003 inches of Al, which is sufficient to stop completely the beta-particles from UX1, so that the ionization in the electroscope is entirely due to the beta-particles from UX2.

When the correction for absorption of the beta-particles has been made, one finds that the 124 mg of uranyl nitrate is equivalent to 42 mg of uranium element; i.e., 500 disintegrations per second. This produces a drift of 2.22 divisions per minute in the electroscope, so that the number of disintegrations necessary to produce one division deflection of the electroscope is 1.35×10^4.

To check the effect of beta-particle reflection, a cobalt target of standard form and superficial mass 1.4 g/cm^2 was laid over the uranyl nitrate source without otherwise disturbing it. The drift of the electroscope increased to 2.62 divisions/minute. This 18 percent increase in the rate of drift may be assumed to be typical of the effects caused by beta-particle reflection from the body of the target in the cases where this result is most pronounced.

The calculation of absolute yields in Table II is carried out on the assumption that the ionization per beta-particle in the active volume of the electroscope is independent of the energy spectrum of the radioelement emitting the beta-particles. This is, of course, not the case, but a correction for the absorption and specific ionization of the beta-particles would be extremely uncertain; none has been attempted.

The sources of error in the yield measurements are listed below, together with an estimate of the percentage error which may, at worst, be due to each.

1. Secondary emission from target	5 percent
2. Errors in measuring film	5 percent
3. Errors in extrapolation of activity back to zero time	3 percent
4. Reflection of beta-particles from body of target	25 percent
5. Error due to assumption that a given ionization corresponds always to the same number of beta-particles	20 percent

Taking into account all of the possible sources of error, it seems safe to say that the yield values may be relied on to an accuracy of about 50 percent; which, considering that the radioactivity is produced in a thick target by an inhomogeneous beam, is perhaps as accurate a value as is required. These measurements are intended chiefly to serve as a guide to future experiments.

5. Elements in Which Activity Has Been Studied

a. Boron

This element is one of those (B, Mg, Al) in which the phenomenon of artificial radioactivity was discovered.[2] It has since been investigated by several workers.[3] The radioelement produced in boron is N^{13}, according to the reaction $B^{10}(\alpha, n)N^{13}$. This radioelement is also produced in the bombardment of C with protons and deuterons.[4] The half-life values obtained in the present work clustered about the accepted value[5] of 10.3 minutes. The yield has been measured (Table II).

b. Nitrogen

The reaction $N^{14}(\alpha, n)F^{17}$ has been known for some time.[6] In the present work, the half-life was measured in samples collected by recoil on a Pt or Ta sheet bombarded in air. The values obtained fluctuated between 62 and 69 seconds, the average of six measurements being 64 ± 6 seconds. It was thought that perhaps the active F^{17} was escaping from the metal foil on which it had been collected, resulting in a lower value of the half-life, so that samples were measured alternately outside the electroscope and inside the ionization chamber of the electroscope itself. No trend in the values of the half-life so obtained

[2] Curie and Joliot, Comptes rendus **198**, 254 (1934).

[3] Curie and Joliot, J. de phys. (7) **5**, 153 (1934); Comptes rendus **198**, 559 (1934); Int. Conf. Phys. London **I**, 78 (1934); Comptes rendus **200**, 2089 (1934); Alichanow, Alichanian, and Dzelepow, Zeits. f. Physik **93**, 350 (1935); Ellis and W. J. Henderson, Proc. Roy. Soc. **A146**, 206 (1934); Fahlenbrach, Zeits. f. Physik **94**, 607 (1935); Meye, Zeits. f. Physik **105**, 232 (1937).

[4] Cockcroft, Gilbert, and Walton, Proc. Roy. Soc. **A148**, 225 (1935); Crane, Lauritsen, and Harper, Science **79**, 234 (1934); M. C. Henderson, Livingston, and Lawrence, Phys. Rev. **45**, 428 (1934).

[5] Allison, Proc. Camb. Phil. Soc. **32**, 179 (1936).

[6] Wertenstein, Nature **133**, 564 (1934); Danysz and Zyw, Acta Phys. Polonica **3**, 485 (1934); Haxel, Zeits. f. Physik **93**, 400 (1935); Fahlenbrach, reference 3; Meye, reference 3.

TABLE I. *Comparative values of half-lives of some radioelements.*

ACTIVE ISOTOPE	REACTION	HALF-LIFE	AUTHORITY
F^{17}	$N^{14}(\alpha,n)F^{17}$	1.07±0.1 min.	Present work
"	"	1.2±0.1 min.	Meye[3]
"	"	1.25±0.1 min.	Haxel[6]
"	"	1.1±0.1 min.	Ellis and Henderson[3]
"	"	1.3 min.	Danysz and Zyw[6]
"	"	1.2 min.	Wertenstein[6]
"	$O^{16}(d,n)F^{17}$	1.16 min.	Newson[11]
"	$O^{16}(p,\gamma)F^{17}$	1.28±0.1 min.	DuBridge *et al.*[23]
Al^{28}	$Mg^{25}(\alpha,p)Al^{28}$	2.57±0.08 min.	Present work
"	"	2.3±0.1 min.	Meye[3]
"	"	2.75±0.33 min.	Curie and Joliot[3]
"	"	3.0±0.2 min.	Alichanow *et al.*[3]
"	"	2.1±0.2 min.	Fahlenbrach[9]
"	"	2.1±0.2 min.	Ellis and Henderson[3]
"	$Al^{27}(d,p)Al^{28}$	2.60±0.08 min.	McMillan and Lawrence[11]
"	$Al^{27}(n,\gamma)Al^{28}$	2.3 min.	Amaldi *et al.*[22]
P^{30}	$Al^{27}(\alpha,n)P^{30}$	2.55±0.05 min.	Present work
"	"	2.9±0.087 min.	Meye[3]
"	"	2.89±0.14 min.	Fahlenbrach[3]
"	"	3.2±0.1 min.	Ellis and Henderson[3]
"	"	3.25±0.2 min.	Alichanow *et al.*[3]
"	"	3.25 min.	Curie and Joliot[3]
"	$S^{32}(d,\alpha)P^{30}$	3±0.1 min.	Sagane[14]
"	$P^{31}(\gamma,n)P^{30}$	3.0±0.2 min.	Bothe and Gentner[19]
Cl^{34}	$P^{31}(\alpha,n)Cl^{34}$	32±1 min.	Present work
"	"	40±5 min.	Frisch[13]
"	$S^{33}(d,n)Cl^{34}$	33±1 min.	Sagane[14]
K^{38}	$Cl^{35}(\alpha,n)K^{38}$	7.65±0.1 min.	Present work
"	"	7.75±0.15 min.	Hurst and Walke[15]
Mn^{56}	$Cr^{53}(\alpha,p)Mn^{56}$	2.66±0.25 hours*	Present work
"	$Mn^{55}(n,\gamma)Mn^{56}$	2.5 hours	Amaldi *et al.*[22]
Cu^{62}	$Co^{59}(\alpha,n)Cu^{62}$	10.0±0.1 min.	Present work
"	$Cu^{63}(n,2n)Cu^{62}$	10.0 min.	Pool *et al.*[27]
"	"	10.5±0.5 min.	Heyn[27]
"	$Cu^{63}(\gamma,n)Cu^{62}$	10.5±0.3 min.	Bothe and Gentner[19]
Cu^{61}	$Ni^{58}(\alpha,p)Cu^{61}$	3.4±0.1 hours	Present work
"	$Ni^{60}(d,n)Cu^{61}$	"	Thornton[29]
Zn^{63}	$Ni^{60}(\alpha,n)Zn^{63}$	34±3 min.	Present work
"	$Zn^{64}(\gamma,n)^{63}$	38±1.3 min.	Bothe and Gentner[19]
Ga^{68}	$Cu^{65}(\alpha,n)Ga^{68}$	68±4 min.	Present work
"	$Ga^{69}(\gamma,n)Ga^{68}$	60±2.5 min.	Bothe and Gentner[19]
Br^{78}	$As^{75}(\alpha,n)Br^{78}$	6.3±0.1 min.	Present work
"	"	6.3 min.	Snell[30]
"	$Br^{79}(\gamma,n)Br^{78}$	5±0.5 min.	Bothe and Gentner[19]

* Counter value.

was observed. The value given here for the half-life of F^{17} is somewhat lower than current values (see Table I).

c. Magnesium

The situation in this element is more complicated. The possible reactions leading to artificial radioelements on the bombardment of Mg with alpha-particles are

$$Mg^{25}(\alpha, p)Al^{28}, \quad (1)$$
$$Mg^{24}(\alpha, n)Si^{27}, \quad (2)$$
$$Mg^{26}(\alpha, p)Al^{29}. \quad (3)$$

It is to be expected that Al^{28} and Al^{29} emit negative electrons, and Si^{27} is a positron emitter. Reaction (1), above, has been verified by many

workers,[7] but the results concerning the other two reactions are somewhat divergent. Ellis and W. J. Henderson[8] find that both reaction (2) and reaction (3) take place when Mg is bombarded with the alpha-particles from radon in equilibrium with its decay products. By counting positive and negative electrons separately in a magnetic field, they determined that the period of Si^{27} was from 6 to 7 minutes, and that of Al^{29} about 11 minutes. Meye,[3] on the other hand, found that the total decay curve of Mg bombarded with alpha-particles from a similar but weaker source could be analyzed into only two exponentials, of which one corresponded to the decay of Al^{28} from reaction (1), while the other, of half-life 6.6 minutes, was attributed to Si^{27}. This latter result confirms that of Fahlenbrach[9] and Eckardt.[10]

It is obvious that chemical separation and counting of electrons separated according to charge are required to decide this problem. The matter has not been settled satisfactorily in the work reported here, but it is clear from the total decay curve of Mg bombarded with alpha-particles (Fig. 2) that at least three different periods seem to be present.

By measuring the decay of an activated Mg sample immediately after a short bombardment (30 seconds), we have determined the half-life of Al^{28} as 154 ± 5 seconds, in good agreement with current values[11] (see Table I). The thick-target yield of reaction (1) has also been determined for a target subjected to a short bombardment.

d. Aluminum

The production of P^{30} by the reaction $Al^{27}(\alpha, n)P^{30}$ is well known.[3] The half-life obtained by us for P^{30} is 2.55 ± 0.05 minutes,[12] which is considerably lower than the accepted values. A typical decay curve of activated Al is shown in Fig. 3. While the initial activity of the P^{30} is

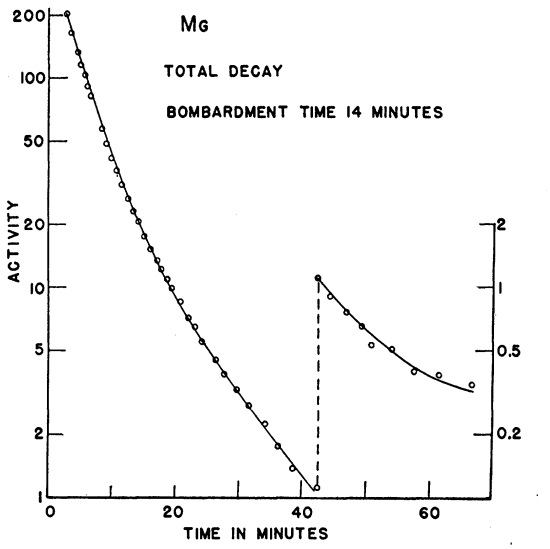


FIG. 2. Total decay curve of Mg bombarded for 14 minutes.

enormous (about 3×10^4 times the background of the electroscope), it is important to be sure that there is present in the target no contaminant which could appreciably shorten the life. The only two contaminants which exhibit activity enough at our bombarding energies to affect the observed life of P^{30} are F^{17} and Al^{28}. Because the former might be formed in considerable strength by recoil from the nitrogen of the air, the targets were bombarded in vacuum; and the chance that Al^{28} would be formed by deuteron contamination of the alpha-particle beam (leading to the reaction $Al^{27}(d, p)Al^{28}$) is considered exceedingly remote in view of the quite complete separation of the H_2^+, D^+ and He^{++} peaks in the cyclotron.[1] The crucial test for the presence of Al^{28}, however, is to determine the sign of the beta-particles emitted from bombarded Al, since Al^{28} emits negative electrons and P^{30} positrons. The beta-particles emitted from the bombarded Al proved to be wholly positive, so that the half-life of P^{30} is, in fact, considerably shorter than has been previously supposed. See Table I.

The yield of the reaction producing P^{30} has been measured. See Table II.

e. Phosphorus

Frisch[13] early discovered that a radioelement which he identified as Cl^{34} was formed in the

[7] Curie and Joliot, reference 3. Alichanow *et al.*, reference 3. Danysz and Zyw, reference 6. Ellis and W. J. Henderson, reference 3.

[8] Ellis and W. J. Henderson, Proc. Roy. Soc. **A156**, 358 (1936).

[9] Fahlenbrach, Naturwiss. **23**, 288 (1935); Zeits. f. Physik **96**, 503 (1935).

[10] Eckardt, Naturwiss. **23**, 527 (1935).

[11] Newson, Phys. Rev. **48**, 790 (1935); McMillan and Lawrence, Phys. Rev. **47**, 343 (1935).

[12] Ridenour, W. J. Henderson, M. C. Henderson, and White, Phys. Rev. **51**, 1013(A) (1937).

[13] Frisch, Nature **22**, 434 (1934).

reaction $P^{31}(\alpha, n)Cl^{34}$. It emits positrons. Frisch's value of the period (40 ± 5 minutes) was improved by Sagane,[14] who found 33 ± 1 minutes for the same radioactive isotope formed in the bombardment of S with deuterons. Our value of the period, 32 ± 1 minutes, is in good agreement with the latter. The thick-target yield has been measured (Table II).

f. Chlorine

K^{38} is produced in the bombardment of chlorine with alpha-particles.[15] Its half-life is 7.65 ± 0.1 minutes. The absorption in Al of the positrons emitted by this radioelement has been measured (Fig. 4); the mass range is about 1.1 g/cm², which indicates, according to Feather's rule,[16] an upper limit of the beta-ray spectrum near 2.3 Mev.

A crude chemical identification of the active isotope was made by dissolving in water the active surface layer of a slab of bombarded rocksalt, adding a little KCl, and precipitating potassium with sodium cobaltinitrite. The potassium cobaltinitrite precipitate carried with it all the activity.

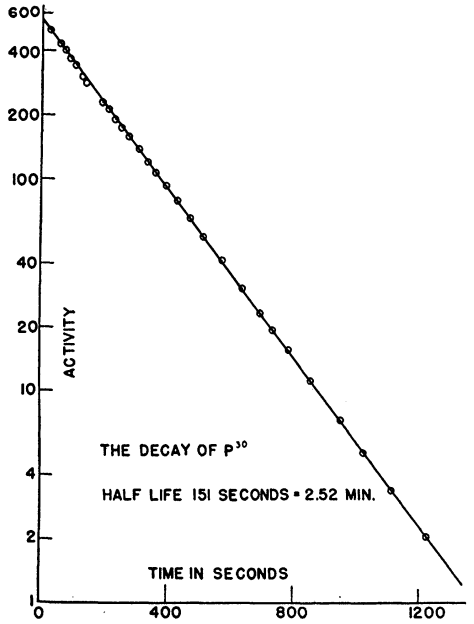


FIG. 3. The decay of P^{30}.

[14] Sagane, Phys. Rev. **50**, 1141 (1936).
[15] Hurst and Walke, Phys. Rev. **51**, 1033 (1937); W. J. Henderson, Ridenour, White, and M. C. Henderson, Phys. Rev. **51**, 1107 (1937).
[16] Feather, Phys. Rev. **35**, 1559 (1930).

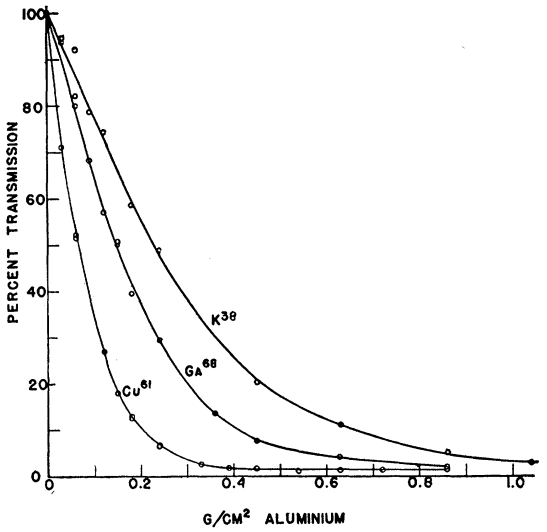


FIG. 4. Absorption in Al of the positrons emitted by K^{38}, Cu^{61} and Ga^{68}.

The first attempt made to measure the yield of this reaction, using a rocksalt target, failed because of the poor conductivity of the salt. A steady deflection of the beam-measuring galvanometer could not be maintained; instead, the target would charge up for a period of perhaps one second, then discharge itself suddenly through the galvanometer, producing a kick in the beam record. This difficulty was overcome by sputtering an extremely thin layer of gold on the surface of the salt and connecting this layer to the galvanometer. The stopping power of the quite transparent gold layer for the alpha-particles of the beam has been estimated as being less than one-tenth of a millimeter, and no correction for it has been applied.

g. Vanadium

The situation in this element is not understood at present. V has only one stable isotope, yet in a sample of V of doubtful purity a strong activity of period about 1.2 minutes was accompanied by a considerably weaker one of roughly 67 minutes half-life. The known alpha-particle reactions: (α, n) and (α, p), provide only one possible radioelement, Mn^{54}, since Cr^{54} is known to be stable, and it was at first thought that at least one of the observed periods in V must be due to an impurity in the sample.

Through the kindness of Mr. D. L. Edlund

and the Vanadium Corporation of America, a sample of unusually pure vanadium was secured, the analysis supplied with it stating that it contained

V	99.6 to 99.8 percent
Fe	0.05 percent
Si	0.05 percent
Al	less than 0.05 percent
N	less than 0.001 percent[17]

Oxides remainder.

This pure V showed both the periods which had been observed in the former sample, although the short-lived radioelement was considerably weaker in the pure sample, and the long one somewhat stronger. This last result makes it appear that the short period activity may be due to an impurity, but the only impurities known to us which could give a comparable period are N(F^{17}), Al(P^{30}), and Mg(Al^{28}). Mg is excluded as a possibility because of the fact that the particles emitted by the short period radioelement produced in V are positrons, and neither N nor Al is present in the sample in sufficient quantity to explain the observed intensity of the short period. Br impurity has not been mentioned as a possibility because of the fact that the short period activity induced in Br, though of the right period (Section 5, *m*), is much too weak to explain the observed activity. Fe, Si, and O are quite inactive relative to the intensities dealt with in this paper (Section 6).

There remains the possibility that the long period activity is due to Cu. (Section 5, *k*). The Cu impurity responsible for the observed activity would have to be present to about 10 percent; this seems out of the question.

One may conclude that of the two activities found in V bombarded with alpha-particles one is due to Mn^{54}. The other is due either (a) to some extremely activable impurity which has been overlooked in the present work, or (b) to Mn^{52} formed from an undiscovered stable isotope V^{49}, or (c) also to Mn^{54}. Mn^{54} finds itself between the isobars Cr^{54} and Fe^{54}, so that it has the alternatives of positron or electron decay leading to a stable isotope. In Cu^{64}, the only similar case where both positrons and electrons are known to be emitted by a single isotope,[18] the half-lives for positron and electron decay are the same, but this is not necessarily true for all such cases. Mn^{54} may, then, show the sort of nuclear isomerism exhibited by Br^{80}, in which two different half-lives exist for the emission of the same sort of particles (negative electrons, in the case of Br^{80}) from a given nuclear species;[19]

[17] By vacuum fusion method.

[18] Van Voorhis, Phys. Rev. **50**, 895 (1936).

[19] Bothe and Gentner, Zeits. f. Physik **106**, 236 (1937).

TABLE II. *Summary of data on radioactive isotopes.*

Bombarded Isotope	Relative Abundance,[25] Percent	Reaction	Radioactive Isotope	Sign of β-Particles	Half-Life	λ Disintegration Constant, min.$^{-1}$	$(dN/dt)_{t=0}$ Initial Intensity Corrected to 10^{13} α-Particles Effective at Zero Time		$No=\frac{(dN/dt)t}{\lambda}=0$ Number of Active Atoms at Zero Time for 10^{13} Effective α-Particles		$Y=(No/A)\times10^{-13}$ Yield Per Incident α-Particle Corrected to Target Composed Entirely of Isotope Concerned
							Div./Min.	Microcuries	Total Divisions	Active Atoms	
B^{10}	20	(α,n)	N^{13}	+	10.3 min.	0.0673	46	0.28	684	4.26×10^{6}	4.6×10^{-6}
N^{14}	99.6	(α,n)	F^{17}	+	1.07 min.	0.648	—	—	—	—	—
Mg^{25}	11.5	(α,p)	Al^{28}	−	2.56 min.	0.271	627	3.8	2310	3.13×10^{7}	2.7×10^{-5}
Al^{27}	100	(α,n)	P^{30}	+	2.55 min.	0.272	3175	19.3	11700	1.59×10^{8}	1.6×10^{-5}
P^{31}	100	(α,n)	Cl^{34}	+	32 min.	0.0216	55	0.335	2540	3.44×10^{7}	3.4×10^{-6}
Cl^{35}*	76	(α,n)	K^{38}	+	7.65 min.	0.0906	76*	0.46*	840*	1.14×10^{7}*	3.0×10^{-6}¶
Cr^{50}(?)	4.9	(α,n)	Fe^{53}(?)	+	8.9 min.§	0.0779	6.8	0.0415	88	1.19×10^{6}	2.2×10^{-6}(?)
Cr^{53}	10.4	(α,p)	Mn^{56}	−	160 min.§	4.33×10^{-3}	—	—	23‖	3.10×10^{5}	3.0×10^{-7}
Ni^{58}	67.3	(α,p)	Cu^{61}	+	3.4 hrs.	3.42×10^{-3}	1.46	0.0089	427	5.80×10^{6}	8.6×10^{-7}
Co^{59}	99.8	(α,n)	Cu^{62}	+	10.0 min.	0.0693	125	0.762	1800	2.44×10^{7}	2.4×10^{-6}
Ni^{60}	26.9	(α,n)	Zn^{63}	+	34 min.	0.0204	1.23	0.0075	60	8.14×10^{5}	3.0×10^{-7}
Cu^{63}	70	(α,n)	Ga^{66}	+	9.4 hrs.	1.23×10^{-3}	—	—	138‡	1.87×10^{6}	2.7×10^{-7}
Cu^{65}	30	(α,n)	Ga^{68}	+	68 min.	0.0102	3.41	0.0208	334	4.53×10^{6}	1.5×10^{-6}
As^{75}	100	(α,n)	Br^{78}	+	6.3 min.	0.110	30.3	0.185	275	3.73×10^{6}	3.7×10^{-7}
Br^{79}†	50	(α,n)(?)	Rb^{82}	+	1.5 min.	0.462	16.6†	0.101†	36†	4.88×10^{5}†	2.0×10^{-7}¶
Br^{81}†	50		Rb^{84}	+(?)	9.8 min.	0.0708	1.8†	0.011†	25.4†	3.44×10^{5}†	1.4×10^{-7}¶

* Rocksalt target.
† NaBr target.
‡ Computed from ratio $Ga^{66}/Ga^{68}=19.8/48$, corrected to infinite bombarding time. Ratio measured in separate experiment.
§ Counter values.
‖ Computed from ratio $Mn^{56}/Fe^{53}=26/100$, corrected to infinite bombarding time. Ratio measured in separate experiment.
¶ Yield has been multiplied by 2 to take account of Na in target.

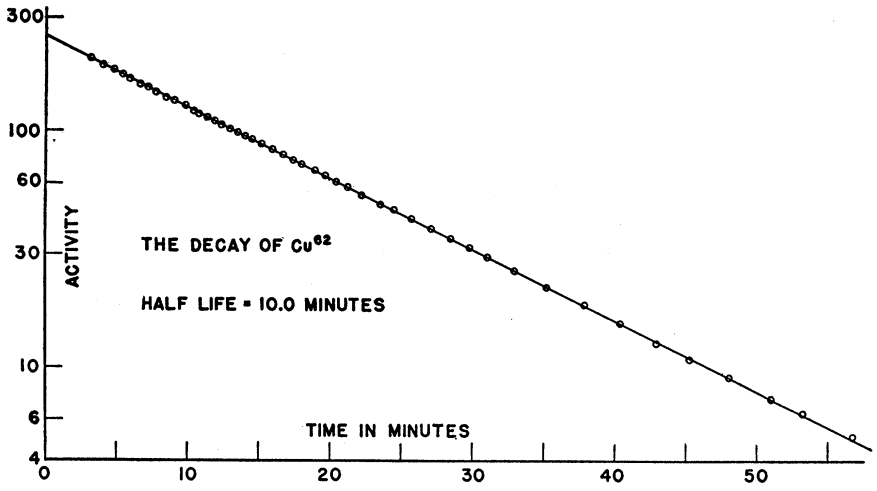


FIG. 5. The decay of Cu^{62}.

it may also be the sort of positron and electron emitting isomer adduced by Pool and others[20] to explain their results on the radioactive isotopes of silver. An attempt made in the present work to determine the sign of the electrons emitted by the long period activity in V bombarded by alpha-particles did not yield definite results.

h. Chromium

The results obtained on bombarding this element with alpha-particles have already been reported briefly.[21] Two activities are found, of which the half-lives are 8.9 ± 0.3 minutes and 160 ± 15 minutes. The latter emits negative electrons, is an isotope of Mn, and is almost certainly to be identified with the Mn^{56} which has been formed by the bombardment of Mn, Fe, or Co with neutrons.[22] The alpha-particle reaction responsible for the production of this radioelement is then $Cr^{53}(\alpha, p)Mn^{56}$. The half-life of Mn^{56} has been given by Fermi[22] as 2.5 hours.

The short-lived radioelement (8.9 minutes) produced in Cr by alpha-particle bombardment emits positrons, and is an isotope of Fe. In an earlier communication,[21] we attributed this activity to Fe^{55}, but this assignment now seems doubtful in view of the results of DuBridge.[23] DuBridge finds that a radioelement isotopic with Fe is formed in the bombardment of Mn with protons; this radioelement has a half-life of 90 minutes. Since Mn has only one stable isotope, the reaction is presumably $Mn^{55}(p, n)Fe^{55}$. Additional weight is given this identification by the fact that DuBridge finds a sharply defined threshold for the reaction producing the 90-minute radioelement at a proton energy of 2.5 Mev; this is a characteristic of the (p, n) reaction.[24]

There remains the possibility that the 8.9 minute activity reported by us in Cr bombarded with alpha-particles is due to Fe^{53}, formed in the reaction $Cr^{50}(\alpha, n)Fe^{53}$. In the absence of other data, we earlier preferred[21] to attribute it to $Cr^{52}(\alpha, n)Fe^{55}$ for the reason that the relative abundances of the Cr isotopes seem to favor that interpretation. The isotopic constitution of Cr is:[25]

Cr^{50}	4.9 percent
Cr^{52}	81.6 percent
Cr^{53}	10.4 percent
Cr^{54}	3.1 percent.

No trace of any long period activity was found in the Fe separated chemically from Cr bombarded with alpha-particles, so that if DuBridge's identification of Fe^{55} is correct, the probability of the reaction $Cr^{53}(\alpha, n)Fe^{55}$ is very small at our alpha-particle energy.

[20] Pool, Cork, and Thornton, Phys. Rev. **52**, 380 (1937).
[21] W. J. Henderson and Ridenour, Phys. Rev. **52**, 40 (1937).
[22] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, and Segrè, Proc. Roy. Soc. **A149**, 522 (1935).
[23] DuBridge, Barnes and Buck, Phys. Rev. **51**, 995 (1937).
[24] DuBridge, private communication.
[25] Hahn, Ber. d. D. Chem. Ges. **70**, 1 (1937).

It should be mentioned that since no stable isotope Mn^{53} has thus far been found,[25] the assignment of the 8.9 minute activity to Fe^{53} requires one of the following alternatives: 1. Mn^{53} is stable, but very rare, or 2. The positron decay of Mn^{53} into Cr^{53} is of so short or so long a life as to have been overlooked in the present work, or of a life so near to 8.9 minutes that the measurements indicated a single period, or 3. Mn^{53} decays to Cr^{53} by K electron capture.

The chemical separation of Cr, Mn, and Fe from bombarded Cr targets was performed for us by Mr. R. C. Newton, to whom we are very grateful. The bombarded Cr was dissolved in 5 ml of 3 N HCl, and 15 mg Fe (as $FeCl_3$), 12 ml concentrated H_2SO_4, and water to make 100 ml were added. A small amount of $KMnO_4$ was added to oxidize the possible Fe^{++} and to supply the required inactive Mn. The solution was cooled to 10° and 6 percent cupferron added in excess. The solution was filtered by suction after several minutes settling, and the Fe precipitate washed with cold 10 percent H_2SO_4 containing 0.2 g cupferron per 100 ml. The iron preciptate was removed to the counter and was found active, its activity decaying with a half-life of 8.9 minutes.

Mn was separated from Cr in the filtrate by addition of Na_2O_2. Oxidation of the residual cupferron yielded tar-like products which were collected on filtration along with the Mn. The Mn precipitate emitted negative electrons, its activity decaying with a half-life of 160 minutes.

Cr was separated from the filtrate of the Mn separation by precipitation as $PbCrO_4$. The Cr precipitate was inactive.

i. Cobalt

The strong activity produced in Co by alpha-particle bombardment decays with a single period[21, 26] of 10.0±0.1 minutes (Fig. 5); positrons are emitted. Co has only one abundant isotope,[25] Co^{59}, and the agreement of the period found in the present work with that ascribed to Cu^{62} by various workers[27] makes it seem very probable that the reaction involved here is $Co^{59}(\alpha, n)Cu^{62}$. The yield of this reaction has been measured (Table II).

j. Nickel

Two radioelements have been produced in Ni by bombardment with alpha-particles, according to the reactions[26, 28]

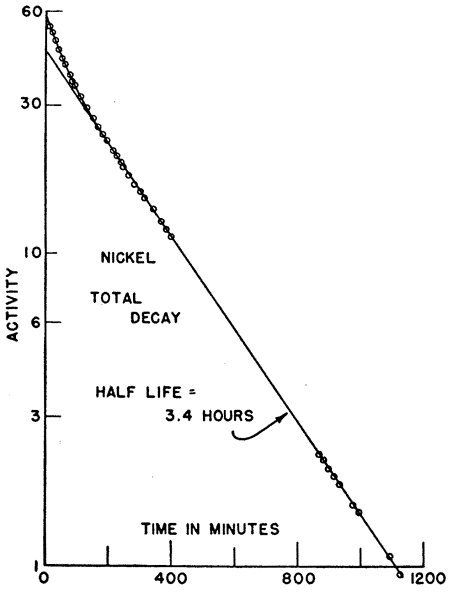


FIG. 6a. Total decay curve of Ni bombarded for 2¾ hours.

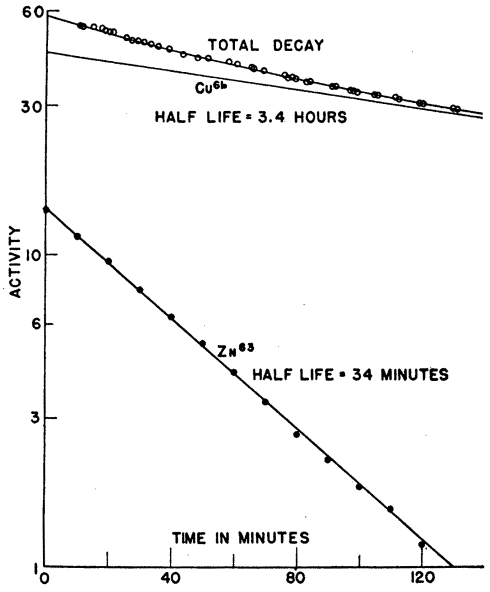


FIG. 6b. Early part of Ni decay curve, showing curves subtracted to obtain period of Zn^{63}.

[26] Ridenour and W. J. Henderson, Phys. Rev. **51**, 1102 (1937).

[27] Heyn, Nature **138**, 723 (1936); Physica **4**, 160 (1937); Pool, Cork, and Thornton, Phys. Rev. **51**, 890 (1937); Bothe and Gentner, reference 18.

[28] Ridenour and W. J. Henderson, Phys. Rev. **52**, 139 (1937).

$Ni^{58}(\alpha, p)Cu^{61}$ and $Ni^{60}(\alpha, n)Zn^{63}$.

Both emit positrons. The absorption in Al of those from Cu^{61} has been measured (Fig. 4); the mass range is about 0.40 g/cm², so that the upper limit of the energy spectrum computed from Feather's rule is 0.94 Mev. The half-life of Cu^{61} is 3.4±0.1 hours; both the half-life and the upper limit of the beta-ray spectrum agree with the values determined by Thornton[29] for the same radioelement obtained in the bombardment of Ni with deuterons.

The 3.4-hour radioelement was shown to be an isotope of Cu by the following procedure: Bombarded nickel foil was dissolved in nitric acid and small amounts of inactive Cu and Zn added. Copper was separated as CuS by bubbling H_2S through the acid solution. The filtrate was made alkaline with ammonia, and the Ni and Zn present precipitated together by passing H_2S through the solution. The Cu precipitate alone was active, the Ni+Zn precipitate not showing perceptible activity 2½ hours after the cessation of the bombardment. These chemical experiments were made on a sample bombarded with particles which had passed through the platinum exit window of the cyclotron and therefore had a mean energy of about 7.5 Mev; under these conditions the amount of Zn^{63} formed is very small,[28] so that the activity due to it could not be detected 2½ hours after the bombardment.

Mr. R. C. Newton performed a check on our identification of the chemical nature of the activity by dissolving the CuS precipitate mentioned above in HCl and electrolyzing out the Cu from acid solution. The activity was found in the electrolytic copper.

The decay of Zn^{63} has never been observed in the absence of Cu^{61} (Fig. 6), so that the period has been determined by the somewhat unreliable method of subtraction. The half-life thus obtained is 34±3 minutes, in fair agreement with the value of 38 minutes attributed by Bothe and Gentner[19] to Zn^{63} formed in the reaction $Zn^{64}(\gamma, n)Zn^{63}$.

The yields of the two reactions which take place in the alpha-particle bombardment of Ni have been measured; the results are found in Table II.

[29] Thornton, Phys. Rev. **51**, 893 (1937).

k. Copper

The two stable isotopes of this element give rise to two radioactive Ga isotopes by the (α, n) reaction[21, 26] as follows:

$Cu^{63}(\alpha, n)Ga^{66}$ and $Cu^{65}(\alpha, n)Ga^{68}$.

Both emit positrons. Ga^{68} has a half-life of 68±4 minutes, as measured in a target bombarded for 15 minutes, the decay being corrected for the small amount of Ga^{66} present. The absorption of the positrons from Ga^{68} has been measured (Fig. 4); the mass range is about 0.86 g/cm² of Al, indicating an upper spectral limit near 1.85 Mev. Ga^{68} has been produced by Bothe and Gentner[19] in the reaction $Ga^{69}(\gamma, n)Ga^{68}$.

The half-life of Ga^{66} is 9.4±0.4 hours. Mr. R. C. Newton performed a chemical separation of Cu, Zn, and Ga which demonstrated that both the radioelements produced in Cu by alpha-particle bombardment are isotopes of Ga. Bombarded Cu was dissolved in 2 *N* HNO_3, and some 30 mg each of Ga and Zn were added in the form of chloride solutions. The solution was made up to 50 ml and 6.5 *N* in HCl, and extracted twice with ether previously saturated over 6.5 *N* HCl. The two ether extracts were combined and washed with 50 ml of 6.5 *N* HCl (saturated with ether), and the ether layer separated and evaporated. The Ga was precipitated from the residue as the ferrocyanide. The decay of the Ga precipitate was followed for 6 hours on the electroscope, and it was found that both the 68-minute radioelement and the 9.4-hour radioelement were present in it.

The Cu and Zn were separated from the remaining HCl solution by sulfide precipitation; both precipitates proved to be inactive.

l. Arsenic

Snell[30] has already reported that a radioactive isotope of Br of half-life 6.3 minutes is formed in the bombardment of As with alpha-particles. The activity is presumably due to Br^{78}, formed in the reaction $As^{75}(\alpha, n)Br^{78}$. Our value of the half-life agrees exactly with Snell's; we have measured the thick target yield of this reaction at our bombarding energy (Table II).

[30] Snell, Phys. Rev. **51**, 1011(A) (1937).

m. Bromine

Two short-lived radioelements are formed in the bombardment of Br with alpha-particles. The periods are about 1.5 minutes and 9.8 minutes. Since the (α, p) reaction leads to stable Kr isotopes in the case of both of the known stable Br isotopes, the reactions producing these radioelements are probably

$$Br^{79}(\alpha, n)Rb^{82} \quad \text{and} \quad Br^{81}(\alpha, n)Rb^{84},$$

although chemical identification has not been carried out in this case. Both radioelements emit positrons, and stable Kr isotopes of mass numbers 82 and 84 exist, so that there are no difficulties in the way of this identification. If this assignment is correct, it is not possible from alpha-particle data alone to decide to which isotope each period corresponds. The activities are somewhat weak at our alpha-particle energy; see Table II for the results of the yield measurement.

6. Elements Displaying Weak Activity

No target has ever been found which, when bombarded for 30 minutes with our alpha-particle beam, gave an initial count less than about 400 per minute on a thin-walled (0.1 mm) Dow metal tube counter. Since so many ubiquitous impurities (N, Al, Cu, P) are tremendously activable, we have postponed the careful study of any radioelements produced by alpha-particle bombardment which are not intense enough to give an initial activity of about 4 div./min. on the electroscope used. This corresponds to an initial intensity of roughly 8000 counts/minute on the counter used.

The following targets are in the class of those which exhibit such weak activities: LiF, Be, C, SiO_2, paraffin, Na_2CO_3, S, Mn, Fe, Zn, Ga_2O_3, Se, Ag, Ta, Mo, Cd, In, Sb, Sn, ZrO_2, Pt, Pd, PbI_2, Rb_2CO_3, and Nb.

To demonstrate the tremendous effect of a trace of an activable impurity, we may state that about 60 counts/min. on the thin walled counter used in these experiments would be produced by one part per million of Al in a target. Co and P impurities would each contribute about 1 count per minute per part per million, if the bombarding time were about half an hour. The requirements of target purity imposed by this tremendous activability are so stringent that investigations of weak activities must be conducted only with the greatest caution.

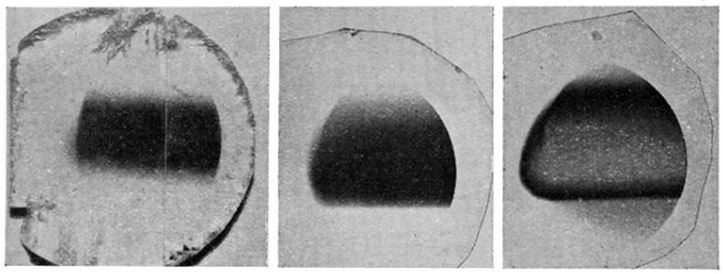
7. Summary

From the results mentioned in Sections 5 and 6, it is evident that the alpha-particle activation of most of the elements up to atomic number 53 has been studied. The noble gases have been omitted because the target-chamber employed was not suited for dealing with gases, and the other elements, namely K,[31] Ca,[31] Sc, Ti,[32] Ge, Sr, Y, Ma, Ru, Rh, Te, have been omitted either because their activity had already been studied or because targets were not readily available. With these exceptions the results given in Table II should represent the only strong activities of moderate lifetime produced by 9 Mev alpha-particles in the elements below iodine.

8. Acknowledgments

Our colleagues, Dr. M. G. White and Dr. M. C. Henderson, have been chiefly responsible for the construction of the Princeton cyclotron, with which these results have been obtained. We are grateful to them for favors too numerous to detail. Professor N. H. Furman of the Department of Chemistry has assisted us greatly by suggesting chemical procedures for the identification of some of the radioelements encountered in our work; some of these, as indicated in the earlier text, have been carried out for us by Mr. R. C. Newton, also of that Department, to whom we owe our thanks. We are indebted to Mr. W. B. Foulk, of the Frick Chemical Laboratory, for placing at our disposal a sample of exceptionally pure chromium, and to Mr. D. L. Edlund and the Vanadium Corporation of America for the sample of extremely pure vanadium mentioned. We are grateful to Professor H. D. Smyth for his continued interest in this work.

[31] Walke, Phys. Rev. **51**, 439 (1937).
[32] Walke, Phys. Rev. **51**, 1011(A) (1937).



a *b* *c*

FIG. 1. Position of beam on target. *a*. Rocksalt sample bombarded for about one minute. Blackened area shows the location of the greater part of the beam. *b*. Record obtained on a piece of film cut roughly to size and shape of target, wrapped in 0.001 inch Al foil, placed in target holder and exposed to alpha-particle beam for about 1/20 second. Sensitivity is greater here than in case of rocksalt; note that traces of beam appear above the rectangular area blackened on the salt. *c*. Film exposed to beam in the same fashion as *b*; exposure time about 2 minutes. Traces of beam now appear below main spot. Note reversal in center, where beam is strongest.