Radioactive Scandium. I*

CARL T. HIBDON,** M. L. POOL, AND J. D. KURBATOV
The Ohio State University, Columbus, Ohio
(Received March 12, 1945)

Sc^{42}: The existence of this radioactive isotope, reported half-life of 13.4 days, has not been confirmed. Sc^{43}: This isotope of 3.92 ± 0.02 hours half-life has been produced by the reactions $Ca(d, n)$, $Ca(\alpha, p)$ and $Ca(p, n)$. Positrons of 1.13 Mev and gamma-rays of 1.65 Mev are emitted. The ratio of the number of positrons to gamma-rays is four. Sc^{44}: One of the isomers of Sc^{44} decays with a half-life of 3.92 ± 0.03 hours. The positrons and gamma-rays have the same energy; namely 1.33 Mev. Three gamma-rays per positron are emitted. Absorption measurements in aluminum and in beryllium indicate characteristic x-rays. The other isomer of Sc^{44} has a half-life of 2.44 days. The genetic relation of the isomers of Sc^{44} has been confirmed.

INTRODUCTION

ARTIFICIAL radioactivity of scandium was first studied by Zyw,[1] who bombarded potassium chloride with alpha-particles of radium C' of 55 mm effective range. A half-life period of three hours was found in the scandium fraction. Zyw concluded that the radioactive isotope formed was Sc^{42} or Sc^{44}. No second period was detected. Later Frisch[2] bombarded calcium fluoride with alpha-particles using 600 millicuries of radon as a source of alpha-particles. A half-life period of 4.4 hours, with a possible error of ten percent, was observed in the scandium precipitate. This half-life period was assigned to Sc^{43}. The protons emitted during the bombardment of calcium to form Sc^{43} have been studied by Pollard and Brasefield,[3] who used ThC' as a source of alpha-particles.

Fermi and his co-workers[4] irradiated calcium fluoride in water for 14 hours with a 600-millicurie radon source mixed with beryllium. No activity was observed in the scandium fraction.

Artificial radioactivity of scandium produced by bombarding scandium itself was first studied by von Hevesy,[5] who observed a long half-life period and assigned it to Sc^{46}.

Among the first studies of scandium made with the aid of the cyclotron may be found those resulting from fast neutron bombardments of scandium.[6] The fast neutrons were obtained by bombarding Li with deuterons. Two half-life periods were observed, 4 hours and 2 days, and were assigned to Sc^{43} and Sc^{44}, respectively. A similar investigation was made by Walke.[7]

A later report included three half-life periods in Sc^{46}, namely, 1.1 hours, 85 days and a period greater than one year.[8] A further search was made by Walke[9] for these three periods. Finally the 85-day period remained as Sc^{46}.

A more extensive study of the various radioactive isotopes of scandium was afterwards made by Walke. A survey was carried out to activate K and Ca by alpha-particle bombardments. Calcium and scandium were in turn respectively bombarded with deuterons and with slow and fast neutrons. Some discrepancies were found in the results obtained from the scandium fractions. Later a more complete study was made in which fast neutron bombardments of titanium and vanadium were included. In this final paper, Walke reported the radioactive isotopes of scandium substantially as they were known to exist when the present research was begun.[10]

* Some of these results have been presented at the Columbus meeting of the American Physical Society, April 30 and May 1, 1943.
** Now at Cornell University, Ithaca, New York.
[1] M. Zyw, Nature **134**, 64 (1934).
[2] O. R. Frisch, Nature **136**, 220 (1935).
[3] E. Pollard and C. J. Brasefield, Phys. Rev. **51**, 8 (1937).
[4] E. Fermi *et al.*, Proc. Roy. Soc. **A149**, 522 (1935).
[5] G. von Hevesy, Nature **135**, 1051 (1935).
[6] M. L. Pool, J. M. Cork and R. L. Thornton, Phys. Rev. **52**, 41, 239 (1937).
[7] H. Walke, Phys. Rev. **52**, 777 (1937).
[8] J. M. Cork and R. L. Thornton, Phys. Rev. **53**, 866 (1938).
[9] H. Walke, E. J. Williams and G. R. Evans, Proc. Roy. Soc., **360**, 171 (1939).
[10] H. Walke, Phys. Rev. **57**, 163 (1940).

APPARATUS AND EXPERIMENTAL TECHNIQUES

All bombardments were carried out with the 42-inch cyclotron of The Ohio State University. The bombarding particles used were 20-Mev alpha-particles, 10-Mev deuterons, 5-Mev protons and fast neutrons produced by the $(Li+d)$ reaction. Irradiations continued from 2 to 16 clock hours.

Decay curves were plotted from data secured from measurements on a Wulf unifilar electrometer with an ionization chamber containing freon gas at a gauge pressure of 20 lb. per square inch. Absorption measurements were also made with the aid of this equipment. For the detection of soft radiations, another Wulf unifilar electrometer and ionization chamber were together inverted. A mechanism was used to adjust open samples of radioactive materials very near the aluminum foil which covers the ionization chamber. The foil has a weight of 0.14 mg/cm^2. Therefore, the ionization chamber was necessarily used at atmospheric pressure.

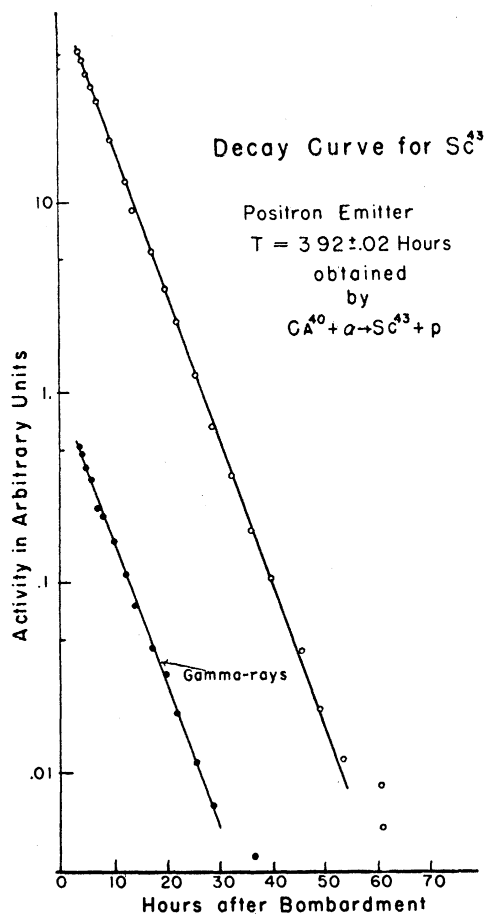

FIG. 1. Sc^{43}: Decay curve showing half-life of 3.92 ± 0.02 hours for both positrons and gamma-rays.

The maximum energy of beta-particles was in most cases found by aluminum absorption measurements. The Sargent formula was used for energies greater than 0.6 Mev and the range-energy relation for energies less than 0.6 Mev.

For the identification of beta-rays a Wilson cloud chamber of 15-cm diameter and a 180° focusing type magnetic spectrometer with a radius of curvature of 16 cm were available.

Gamma-ray energies were measured by absorption in lead and checked by copper and sometimes also by aluminum. A maximum thickness of three inches of lead or of copper was used wherever possible.

In order to obtain the correct absorption coefficient for samples emitting positrons, it was necessary to enclose the sample completely in lead to annihilate all positrons near the sample.

The chemical separations have been made for potassium, calcium and scandium targets. In all cases, the scandium fraction has been separated first by precipitation with oxalic acid and then freed from common impurities by addition of corresponding carriers. The hydrofluoric acid procedure and ammoniacal procedure were employed in purification.

Sc^{42}

A radioactive half-life period of 13.4 days was assigned to Sc^{42} by Walke[10] and was reported to have been produced by the $K^{39}(\alpha, n)$ and $Ca^{40}(\alpha, d)$ reactions. Throughout the present experiments no evidence could be found to indicate the existence of this radioactive period despite repeated efforts to produce and confirm it.

Sc^{43}

A half-life period of scandium varying from 3 to 4.4 hours has often been reported and variously assigned by different experimenters to Sc^{42}, Sc^{43}, or Sc^{44}. The latest assignment was made to Sc^{43} by Walke,[10] who reported a half-life of 4.0 ± 0.1 hours. By aluminum absorption, Walke found two components in the spectrum

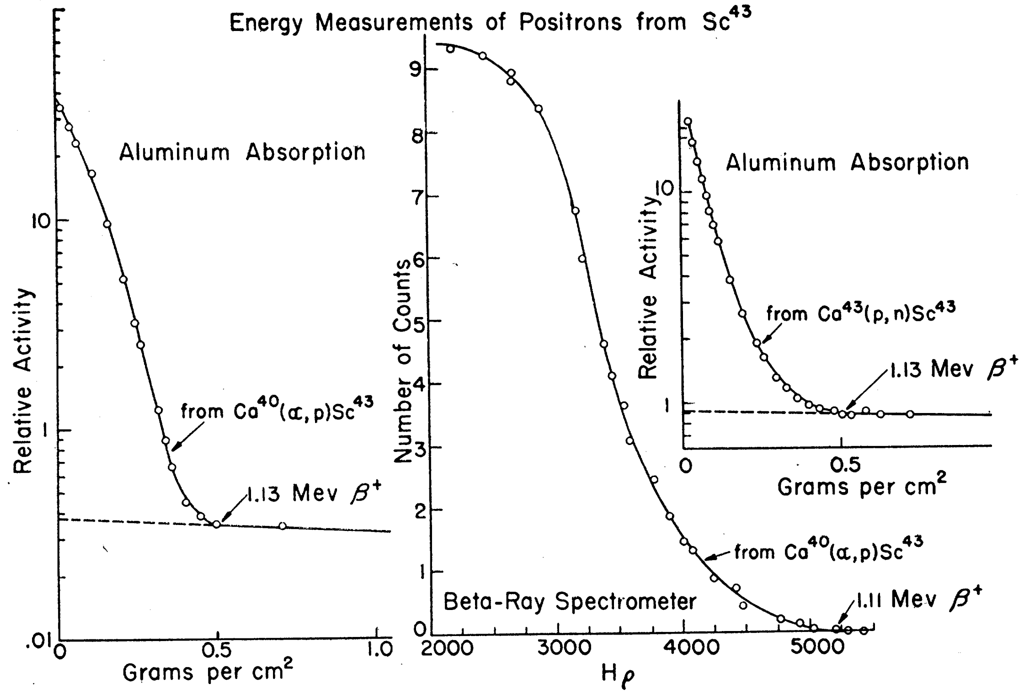

FIG. 2. Sc^{43}: Aluminum absorption and spectrometer measurements of positron spectrum obtained from the $Ca^{40}(\alpha, p)$ and $Ca^{43}(p, n)$ reactions. The end-point is 1.13 Mev.

of Sc^{43} with endpoints at 0.4 Mev and 1.4 Mev (Feather formula). By lead absorption measurements, 1.0 Mev gamma-rays were observed. Sc^{43} was produced by bombarding calcium with deuterons or with alpha-particles.

During the course of the present experiments, Sc^{43} has been rendered extremely active by bombarding calcium with alpha-particles. With this type of bombardment, it is by far the most active isotope of artificially radioactive scandium. Activities ranging up to 30,000 times background have been obtained.

An average half-life of 3.92 ± 0.02 hours was computed from five bombardments. The half-life computed from each bombardment differs very little from this value. A typical decay curve for both the positrons and gamma-rays is shown in Fig. 1.

From aluminum absorption measurements, as shown in Fig. 2, the maximum energy of the positrons was estimated to be 1.13 ± 0.05 Mev (Sargent formula). Measurements made with the magnetic spectrometer indicate a value of 1.11 ± 0.05 Mev.

An energy of 1.65 Mev for the gamma-rays emitted by Sc^{43} has been determined by absorption in lead and in copper. Measurements for lead, as shown in Fig. 3, made with an extremely intense source gave an absorption coefficient of 0.56 cm^{-1}, corresponding to an energy of 1.65 Mev. A very prominent annihilation radiation indicates that more positrons are emitted than 1.65 Mev gamma-rays. From a determination of the ratio of the ionization constant of positrons to that of gamma-rays, it was estimated that one gamma-ray was emitted for every four positrons. The probability that Sc^{43} decays to the ground state of Ca^{43} by positron emission is thus four times that to the excited state by K-electron capture. The presence of x-rays in the difficult region of 3.00A associated with the K-electron capture was not investigated for this isotope.

The detection of the presence of Sc^{43} when produced by proton-bombardment of calcium is complicated by the possible co-production of Sc^{44} and Sc^{48}. However, in the present work the production of Sc^{43} by this type of bombardment has been established as indicated by the half-life, the energy of the positrons and gamma-rays and

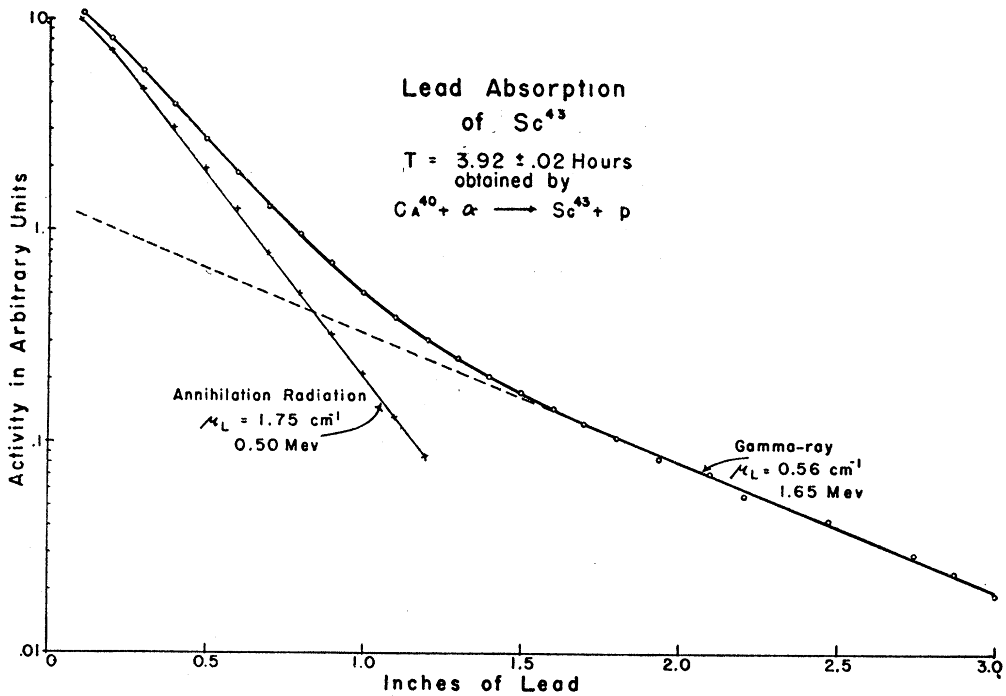

FIG. 3. Sc^{43}: Lead absorption showing gamma-rays of 1.65 Mev.

by the ratio of the intensity of the positrons to that of the gamma-rays.

The detection of the presence of Sc^{43} when produced by deuteron-bombardments of calcium is also complicated by the fact that Sc^{43} and Sc^{44} each have a half-life of 3.92 hours. Nevertheless, the presence of Sc^{43} may be established in the presence of Sc^{44} through their respective beta-ray energies of 1.13 and 1.33 Mev and their gamma-ray energies of 1.65 and 1.33 Mev.

THE ISOMERS OF Sc^{44}

As previously reported, Sc^{44} was thought to have genetically related isomers of 4.1- and 52-hour half-lives.[10,11] The 52-hour period (2.16 days) was reported to decay into the 4.1-hour period by the emission of a 0.25 Mev internally converted gamma-ray, and the 4.1-hour period decayed into Ca^{44} by emitting 1.6 Mev positrons.

A restudy of this isotope shows that the scandium fraction of potassium bombarded with

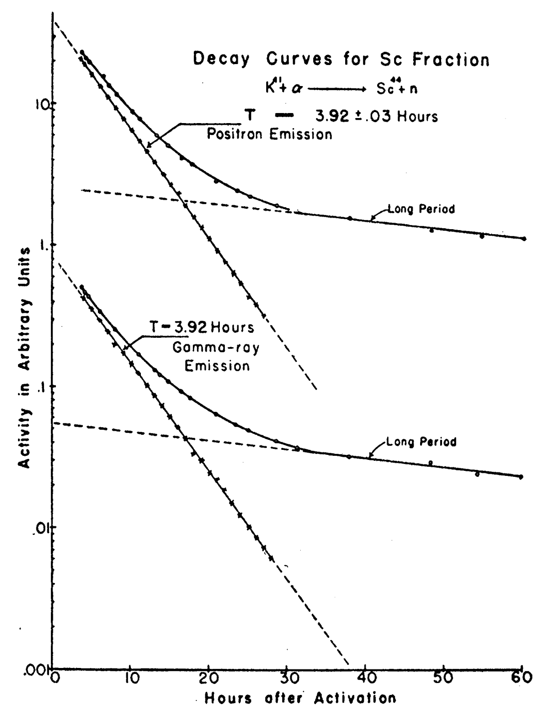

FIG. 4. Sc^{44}: Decay curve showing half-line of 3.92±0.03 hours for both positrons and gamma-rays.

[11] W. E. Burcham, M. Goldhaber and R. D. Hill, Nature **141**, 510 (1938); J. M. Cork and R. L. Thornton, Phys. Rev. **53**, 866 (1938).

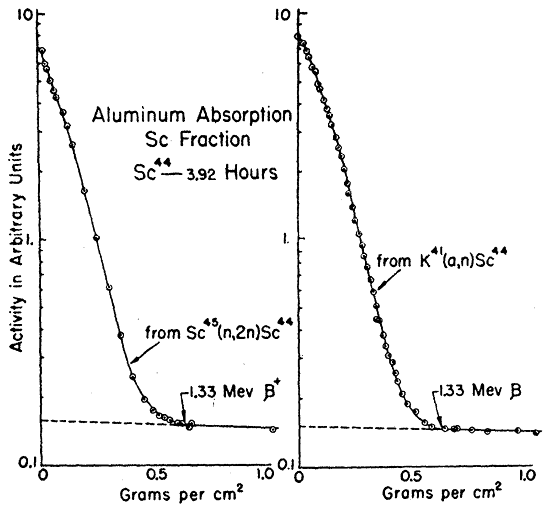

FIG. 5. Sc^{44}: Aluminum absorption measurements of positron spectrum in the 3.92-hour period obtained from the $Sc^{45}(n, 2n)$ and $K^{41}(\alpha, n)$ reactions. The end-point is 1.33 Mev.

alpha-particles yields two main half-life periods, one of 3.92 hours and another of 2.44 days. The same periods were obtained by bombarding scandium with fast neutrons (Li$+d$). By means of these two bombardments Sc^{44} was obtained free of Sc^{43} and Sc^{48}.

The 3.92-Hour Activity

In Fig. 4 a decay curve is shown for an alpha-particle bombardment of potassium. A very similar curve is obtained for a fast neutron bombardment of Sc. The average half-life obtained from several bombardments was found to be 3.92 ± 0.03 hours in each type of bombardment. The original intensity of the period produced by the $K^{41}(\alpha, n)$ reaction was about 3000 times background while the intensity produced by the $Sc^{45}(n, 2n)$ reaction was only 1000 times background. In each case the subtracted decay curve continued for about seven half-lives.

Aluminum absorption measurements were made at various times during the decay. Typical curves are shown in Fig. 5.

An energy of 1.33 Mev for the gamma-rays has been determined by absorption in lead. Measurements, as shown in Fig. 6 for the $K^{41}(\alpha, n)$ reaction, made with a strong source indicate a small amount of annihilation radiation, and consequently more 1.33 Mev gamma-rays are expected than positrons.

Using the relation between the ionization constants of beta-rays and gamma-rays obtained in the study of Sc^{43}, it was estimated that three gamma-rays are emitted per positron. Absorption measurements in aluminum and in beryllium indicated the presence of x-rays between 2.4 and 2.8A. K-electron capture is therefore strongly indicated.

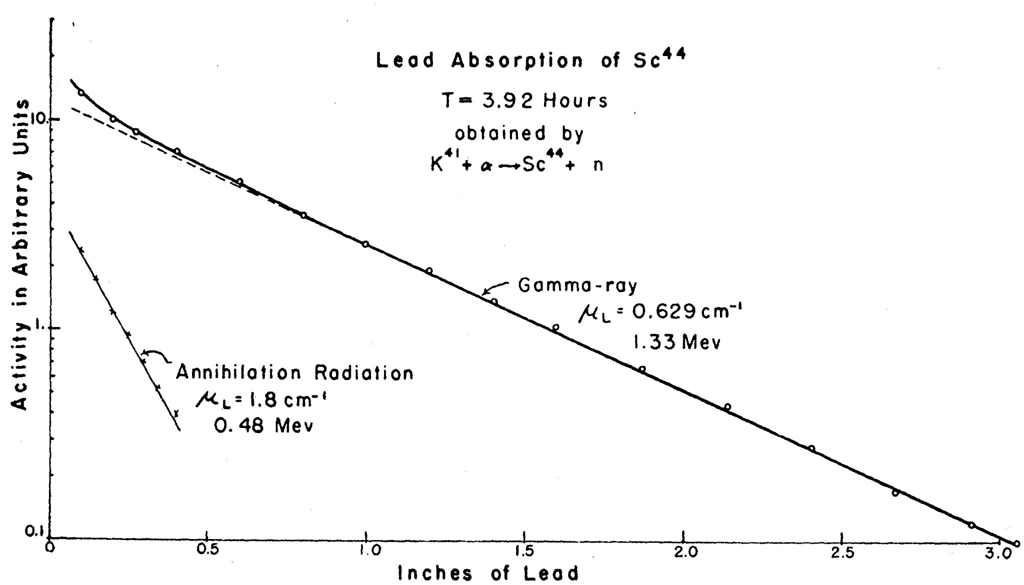

FIG. 6. Sc^{44}: Lead absorption showing gamma-rays of 1.33 Mev in the 3.92-hour period.

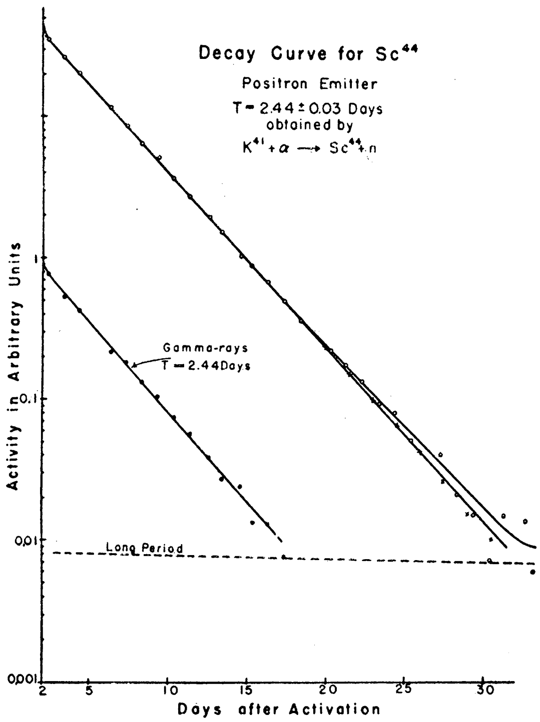

FIG. 7. Sc^{44}: Decay curve showing a half-life of 2.44±0.03 days for both positrons and gamma-rays.

The 2.44-Day Activity

Early during the present work, it was found in bombardments of calcium with deuterons that the gamma-rays of the scandium fraction decayed with a half-life period of 1.83 days rather than with the expected one of 2.16 days.[10] This was reported by Smith[12] and by Mandeville.[13] It remained to be established whether Sc^{44} decayed with a half-life of 2.16 days for the positrons and 1.83 days for the gamma-rays, or whether other radioactive scandium isotopes played an important role.

In order to establish the characteristics of Sc^{44}, it was produced by the reactions $K^{41}(\alpha, n)$ and $Sc^{45}(n, 2n)$. One of the decay curves of the scandium fraction is shown in Fig. 7. From this curve and other similar curves, an average half-life of 2.44 days was observed for both positrons and gamma-rays. The activity could usually be followed for 12 half-lives.

In Fig. 8, similar results are shown for a fast neutron bombardment of scandium. The ratio

[12] Gail P. Smith, Phys. Rev. **61**, 578 (1942).
[13] C. E. Mandeville, Phys. Rev. **62**, 555 (1942); Phys. Rev. **64**, 147 (1943).

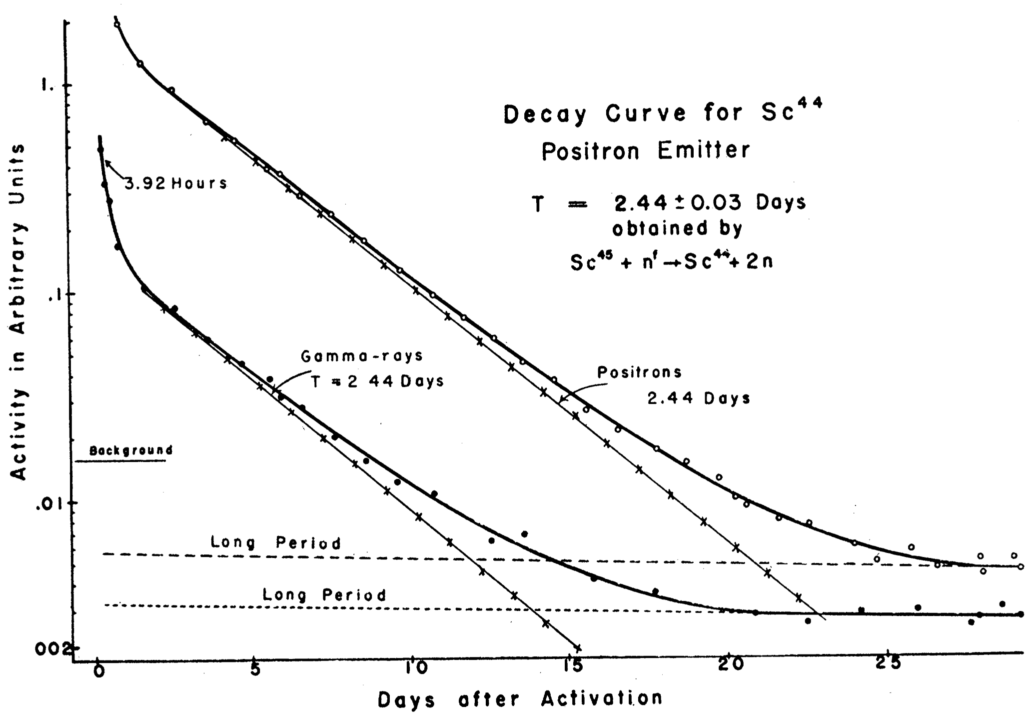

FIG. 8. Sc^{44}: Decay curve showing half-lives of 3.92 hours and 2.44 days for both positrons and gamma-rays obtained by the $Sc^{45}(n, 2n)$ reaction.

of ionizing intensities of the beta-rays to that of the gamma-rays for both bombardments is about 40. This ratio for calcium deuteron bombardments is, however, approximately 15.

From the foregoing data, it therefore appears that deuteron bombardments of calcium produce some other scandium isotope in greater abundance than Sc^{44}. This will be discussed in a later paper.

By aluminum absorption, as indicated in Fig. 9, positrons of maximum energy 1.33 Mev were measured for the $K^{41}(\alpha, n)$ reaction. For comparison the absorption curve for the $Sc^{45}(n, 2n)$ reaction is also included. By absorption measurements made with the inverted electrometer, low energy electrons were observed which are probably due to internally converted gamma-rays of 0.28 Mev energy.

An unconverted gamma-ray was also observed. The energy was 1.33 Mev by absorption in lead, in copper, and in aluminum.

The 1.33 Mev positrons, the 1.33 Mev gamma-ray and the 0.28 Mev internally converted gamma-ray persist throughout the entire decay of the 2.44-day scandium fraction from which it is concluded that the 2.44-day period decays into the 3.92-hour period by emitting the 0.28 Mev gamma-ray. The 1.33 Mev positrons and the 1.33 Mev gamma-rays are, therefore, associated only with the 3.92-hour period.

DuBridge and his co-workers[14] reported the production of the isomers of Sc^{44} by bombardment of calcium with 4 Mev protons. Walke[10] repeated this bombardment with 4 Mev protons and obtained confirmatory results. In the present work no evidence was found to substantiate this

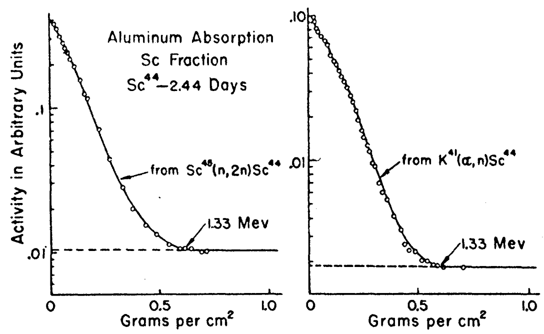

FIG. 9. Sc^{44}: Aluminum absorption measurements of the positron spectrum in the 2.44-day period obtained from the $Sc^{45}(n, 2n)$ and $K^{41}(\alpha, n)$ reactions. The end-point is 1.33 Mev.

reaction although 5 Mev protons were used. However, abundant evidence was found that Sc^{43} was produced.

ACKNOWLEDGMENT

It is a pleasure to express gratitude for the support received from the University Development Fund and from Mr. Julius F. Stone. Very pure scandium was kindly furnished us by Professor L. L. Quill of Michigan State College. Valuable assistance received through the Graduate School is also acknowledged.

[14] L. A. DuBridge, S. W. Barnes, J. H. Buck and C. V. Strain, Phys. Rev. **53**, 447 (1938).