belongs to which period, pictures were taken after 30 and 60 days. After 60 days the 86-kev  $\gamma$ -ray was relatively more intense than at the beginning so that it must be the  $\gamma$ -ray of the 90-day isomeric transition, the 102 kev that of the 32-day transition.

A  $\gamma$ -ray in Te with l=5 and an energy of 102 kev corresponds to a calculated half-life of 2400 days, too large by a factor of 75. On the other hand, the assumption of l = 4 gives a half-life only 1/5000 the observed. The value of 5, therefore, seems quite probable. Using this value, one then calculates that a K/L ratio of 1 is consistently explained by 45 percent electric 25-pole radiation, 55 percent magnetic 2<sup>4</sup>-pole radiation. Similarly, a  $\gamma$ -ray with l=5 and an energy of 86 kev in Te, gives a calculated half-life of 40,000 days,  $400 \times \text{longer}$  than the observed 90 days. However, l=4 gives only 10 minutes as the calculated halflife, and since this is only 1/13,000 the observed value, l=5 seems much to be preferred. The estimated K/L ratio of 0.75, which is probably not wrong by more than 30 percent, then gives for this case also a mixture of 50 percent electric, 50 percent magnetic radiation. The calculated conversion coefficients are large, 2000 and 10,000, respectively; and no unconverted  $\gamma$ -rays could be observed.

#### CONCLUSION

The results discussed above indicate that in many cases of nuclear  $\gamma$ -rays, it is feasible to determine the type of the radiation, electric, magnetic, or both, and the multipole order. Although present formulae for the lifetimes of  $\gamma$ -ray transitions usually indicate a preferable value of the multipole order, in only two or three out of ten cases is the agreement of theory and experiment satisfactory. Therefore, an improvement of the theory on this point would be valuable.

In conclusion, I wish to thank Professor E. O. Lawrence, Professor L. W. Alvarez, and Dr. E. Segrè for their interest and assistance in this work; and Mr. E. Nelson for helpful discussions of the theory. This work has been aided by the financial assistance of the Research Corporation.

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#### PHYSICAL REVIEW

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# Artificial Radioactivity of Ti<sup>45</sup>

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A radioactive isotope possessing a half-life of  $3.08\pm0.06$  hours has been produced by four different types of bombardment. Evidence is presented which indicates that the activity should be assigned to Ti<sup>45</sup>. The four nuclear reactions are:

$Ca^{42} + He^4 \rightarrow Ti^{45} + n^1$	$Sc^{45}+H^2 \rightarrow Ti^{45}+2n^1$
$Sc^{45}+H^1 \rightarrow Ti^{45}+n^1$	$Ti^{46} + n^1 \rightarrow Ti^{45} + 2n^1.$

Nuclear spin considerations indicate that a fifth reaction,  $Ti^{46}(\gamma,n)Ti^{45}$ , is improbable, as was verified experimentally. Analysis of cloud-chamber pictures reveals the maximum positron energy to be 1.2 Mev. On a Sargent diagram, the reaction  $Ti^{45} \rightarrow Sc^{45} + e^+$ , is a permitted one. The half-life of  $3.08 \pm .06$  hours for  $Ti^{45}$  is the weighted average of 3.17, 3.10, and 3.02 hours from Sc(p,n); 3.04 hours from Sc(d,2n); 3.17 hours from  $Ca(\alpha,n)$ ; 3.0 hours from Ti(n,2n).

#### INTRODUCTION

THE elements in the titanium region, shown in Fig. 1, have been quite thoroughly examined for radioactive isotopes by Walke and others. Walke<sup>1</sup> has reported many new radioactive isotopes in potassium, calcium, scandium, titanium, and vanadium, of which the two in scandium,  $Sc^{43}$  (4 hours) and  $Sc^{44}$  (4.1 hours) have

<sup>1</sup>H. Walke and others, Phys. Rev. **51**, 143A, 439, 1011A, 1033 (1937); **52**, 669, 777 (1937); **57**, 163, 171, 177 (1940); Proc. Roy. Soc. **A171**, 360 (1939).

<sup>\*</sup> On sabbatical leave from Bethany College, Bethany, West Virginia.



FIG. 1. Nuclear transmutation chart of the titanium region.

periods close to Ti45 and are of some interest in connection with the present work. The only activities in titanium that have been reported are the isomers of Ti<sup>51</sup> (2.8 minutes and 72 days).<sup>2</sup> Calcium, scandium, and titanium have been subjected to the various bombardments but no reference has been found to the Sc + p reaction which may produce  $Ti^{45}$ . The reactions, Sc+d,  $Ca + \alpha$  and Ti + n (fast) have been reported in the literature. However, the presence of the active isotope Ti<sup>45</sup> has not been reported. In the present work, the first bombardment of scandium oxide with protons, which was followed by a chemical separation of the titanium, showed a three-hour period in the titanium fraction. The observation of this new period aroused our interest not only in assigning this period but in finding the experimental conditions which had prevented previous investigators from detecting this strong activity.

The present work was done on the 42-inch cyclotron at The Ohio State University. Radioactive measurements were made on a Wulf electrometer with an ionization chamber filled with two atmospheres of Freon. A submersive type Geiger-Müller tube was used throughout the chemical procedure to follow the extent of chemical separation.

ACTIVATION OF SCANDIUM WITH PROTONS

When scandium oxide was bombarded with 5-Mev protons for several hours, an isotope exhibiting a very strong half-life of approximately three hours was produced. The first determination of the activity showed it to have an intensity nearly 1000 times the background. The decay has been followed for more than eleven half-lives to an intensity of one-fourth background (Fig. 2). The period obtained from this curve is 3.02 hours. Values determined for other samples bombarded under similar conditions are 3.17 and 3.10 hours.

Nearly all of the activity is due to positron emission. There are a few electrons and some gamma-radiation, as measured on the electrometer, probably due to annihilation radiation. However, the quantity of gamma-radiation associated with this period is very small.

Since scandium has but a single stable isotope, the reaction  $Sc^{45}(p,n)Ti^{45}$  should be the first considered. The formation of Ti45 was later confirmed by other nuclear reactions and other supplementary data.

It has been found<sup>3,4</sup> that certain radioactive elements, even in minute concentrations, may be separated in a very pure form in the absence of a carrier. The basic principle involved in this major distinction from accepted radio-chemical procedure has been extended by one of us (J.D.K.) to work given here on the separation of scandium and titanium and will be described in detail.

The precipitation of Ti<sup>45</sup> in the pure state, without the addition of inactivated titanium from scandium has the advantage that the absence of a large quantity of titanium permits fast separation. This procedure reduces the amount of titanium in the precipitated state which impedes filtration and augments contamination of the titanium through adsorption of active elements. Moreover, the chemical procedures for the separation of scandium and titanium were arranged in such a way that other possible activities which might appear after bombardment could be removed and identified. During chemical separations and purifications, the volume of each fraction was estimated and the activity of ten milliliters measured with the Geiger-Müller counter.

The following elements were considered as

<sup>&</sup>lt;sup>2</sup> H. Walke, Phys. Rev. 51, 1011 (1937); Walke, Williams, and Evans, Proc. Roy. Soc. A171, 360 (1939).

<sup>&</sup>lt;sup>3</sup> J. D. Kurbatov, J. Phys. Chem. **36**, 1241 (1932). <sup>4</sup> J. D. Kurbatov, M. L. Pool, and H. B. Law, Phys. Rev. **59**, 919A (1941).

possible minor impurities: carbon, sodium, magnesium, silicon, phosphorus, potassium, titanium, and iron. In addition, active isotopes arising from calcium and oxygen present in the scandium oxide, and from the copper target, were stabilized and removed.

The chemical procedure after the deuteron bombardment of scandium was similar to that used after the proton bombardment except for the elimination of the different radioactive isotopes formed by deuteron bombardment.

The procedure consisted in dissolving the activated scandium oxide in nitric acid. The resulting solution was then successively evaporated with additions of nitric, of hydrochloric, and of hydrofluoric acids to remove any  $N^{13}$ ,  $O^{15}$  and  $F^{18}$  present.

This treatment was followed, after the addition of one-hundredth gram-ions of sodium, potassium, magnesium, calcium, copper, and cobalt to stabilize the corresponding radioactive isotopes in solution, by the precipitation of scandium and radioactive titanium with excess ammonia. However, no inactivated titanium was added as a carrier. After filtration, the activity of the solution was measured with the G-M immersion tube. The above precipitation of scandium and titanium with stabilizers was repeated if any perceptible activity due to impurities was observed.

There are two procedures commonly used in the separation of scandium and titanium, namely, precipitation of scandium with hydrofluoric acid and with oxalic acid. Both reactions were employed in the present work. The first one was used primarily for the separation of titanium and scandium, while the second was used only for purification.

After the precipitated hydrous oxides of scandium and radioactive titanium were treated with acid, considerable active titanium still remained on the filter paper, even after the scandium was dissolved. The solution was then treated with hydrofluoric acid to precipitate the scandium. After filtration, most of the active substance was found in the solution and was called titanium fraction A.

The scandium precipitate was dissolved and inactive titanium was added for the first time. The scandium was reprecipitated with hydrofluoric acid yielding titanium fraction B and scandium fraction 1.

The titanium that remained on the filter paper after the first filtration was dissolved in acid, inactive scandium was added, and the solution was treated with hydrofluoric acid. This procedure gave titanium fraction C (filtrate) and the practically inactive scandium fraction 2 (precipitate).

The three fractions of titanium were purified separately by the precipitation of titanium with ammonia after stabilizers for the impurities had been added; and the two scandium fractions were dissolved and reprecipitated as the oxalate in the presence of stabilizers for the possible impurities, Fe<sup>59</sup>, P<sup>32</sup>, Zn<sup>63, 65</sup>.

The relative intensities of the activities in the different fractions of titanium and scandium can be seen in Table I. The activities are computed for the same time, a few hours after bombardment.

The activity measured in the three titanium fractions, A, B, C, was found to be the three-hour period. The weak activity of the scandium fractions indicated the efficiency of separation.

To test the efficiency of precipitation of scandium and titanium without the use of a carrier,



FIG. 2. Decay curve for Ti<sup>45</sup>.

the following experiment was performed: Activated scandium oxide (bombardment time, 2 hr.) was dissolved in hydrochloric acid and the solution was diluted to one liter. Only three milliliters of this solution were used, from which the scandium and active titanium were precipitated by ammonia without a carrier. The precipitate contained 7.57 units of activity seven hours after bombardment, and the half-life was determined as 3.11 hours, which indicates the presence of Ti<sup>45</sup>. The filtrate contained 0.5 unit of activity or 6.6 percent. Since the half-life of Ti<sup>45</sup> is 3.08 hours, the decay constant  $\lambda$  is  $6.22 \times 10^{-5}$ . If one accepts arbitrarily that the unit of activity measured on the electrometer is of the order of magnitude of one microcurie, the total weight of Ti<sup>45</sup> precipitated without carrier was found to be 4.45×10<sup>-14</sup> gram.

### ACTIVATION OF SCANDIUM WITH DEUTERONS

Scandium oxide was bombarded with 10-Mev deuterons for four hours, and the active titanium was separated chemically. The activity of this titanium fraction was found to have a period of 3.04 hours, and was followed for five half-lives.

The activity produced in scandium with either protons or deuterons is of such an intensity that it can be followed and the half-life obtained even without chemical separation. This strong activity apparently did not disturb the determination by other investigators of the 85-day period of Sc<sup>46</sup>. Thus, the Ti<sup>45</sup> must have been completely separated chemically from the scandium, even though no attempt was made to separate titanium but only to establish the scandium period.<sup>5</sup> If the filtrate, after separation of scandium, was not tested for activity, it might easily have been discarded.

Thus these investigators confirmed our con-

TABLE I. (Sc+p). Relative activities of titanium and scandium.

FRACTION	RELATIVE ACTIVITY
Ti A	187
Ti B	3.4
Ti C	31
Sc 1	1.58
Sc 2	

<sup>5</sup> H. Walke, Phys. Rev. 52, 669 (1937).

tention that pure radioactive titanium can be separated without a carrier.

Also, the same workers did not observe Ti<sup>45</sup> after bombardment of calcium with  $\alpha$ -particles.<sup>6</sup> The loss of Ti<sup>45</sup> was probably due to the chemical separation of scandium, titanium, and calcium which was performed with the addition of scandium and titanium carriers, according to classical radio-chemistry, and in which the recovered titanium fraction had an activity similar to that of the scandium fraction. This factor indicates the contamination of the titanium fraction by scandium.

## Activation of Calcium with Alpha-Particles

The calcium oxide used in this bombardment was prepared from chalk by several reprecipitations with ammonium carbonate and final conversion to the oxide. The latter was bombarded with 20-Mev alpha-particles for five hours. The titanium fraction obtained after separations of titanium from scandium displayed a half-life of 3.17 hours. This period was followed for four halflives, indicating the reaction  $Ca^{42}(\alpha,n)Ti^{45}$ . One might have expected a very weak activity in titanium because the abundance of  $Ca^{42}$  is only 0.64 percent.

Since the predominating activity from this bombardment was the four-hour period in scandium obtained from an  $\alpha, p$  reaction of Ca<sup>40</sup>, the chemical procedure was arranged with the single purpose of separating Ti<sup>45</sup> in the shortest possible time. Therefore, the usual analytical procedure for separating the elements calcium, scandium, and titanium was disregarded.

The procedure consisted in solution of the bombarded calcium oxides in acid, precipitation of scandium and titanium without carriers, filtration of calcium from scandium and titanium, and purification later from other possible activities as described under the proton and deuteron bombardments.

The filter paper, even with no visible precipitate, had practically all of the scandium and titanium activities. The filtrate, containing calcium, had only a very small percentage of the

<sup>&</sup>lt;sup>6</sup> H. Walke, Phys. Rev. 57, 169 (1940).

total activity as measured on the Geiger-Müller counter, so it was disregarded.

The scandium and titanium precipitate was dissolved in acid and the solution was treated with hydrofluoric acid, after a scandium carrier had been added. The purification of the scandium and titanium fractions was made as described under the (Sc,p) reaction.

# Activation of Titanium with Fast Neutrons and Gamma-Radiation

Titanium oxide was bombarded for six hours with fast neutrons from the Li+d reaction. For this bombardment, commercial TiO<sub>2</sub> was purified by the repeated solution and precipitation of titanium. Analysis of the latter indicated 99.8– 99.9 percent TiO<sub>2</sub>. After bombardment, the weak 3.0-hour activity of the titanium fraction could be followed for only two half-lives, beginning at seven times the activity of the background. These measurements were used to verify the Ti<sup>46</sup>(n,2n)Ti<sup>45</sup> reaction, but were too uncertain to be used in determining the final figure for the half-life of Ti<sup>45</sup>.

With gamma-radiation from the Li+p reaction, on a TiO<sub>2</sub> target, no three-hour activity was obtained. The following nuclear spin considerations suggest an explanation. The angular momentum of Sc<sup>45</sup> has been observed<sup>7</sup> to be  $\frac{7}{2}$ ; and since Ti<sup>45</sup> $\rightarrow$ Sc<sup>45</sup>+ $e^+$  is a permitted transition (as shown below) the angular momentum of the titanium nucleus should also be in the neighborhood of  $\frac{7}{2}$ . But since the Ti<sup>46</sup> nucleus probably has 0 or 1 unit of spin, bombardment with gamma-rays could hardly increase this to  $\frac{7}{2}$ .

### MAXIMUM POSITRON ENERGY

Pictures were taken of the positron tracks in a cloud chamber containing hydrogen at atmospheric pressure and saturated alcohol vapor. The radii of curvature of 887 tracks were measured in



FIG. 3. Positron histogram of Ti<sup>45</sup>.

a magnetic field of 256 gauss. From the spectrum shown in Fig. 3, the maximum positron energy by inspection is 1.24 Mev. When this end point, together with the half-life, is plotted on a Sargent diagram, the point falls on the first curve with the permitted transitions. The upper limit of the positron spectrum from a Fermi plot is 1.26 Mev, and from a Konopinski-Uhlenbeck plot, 1.40 Mev.

If the logarithm of the half-life of Ti<sup>45</sup> is plotted against its atomic number, the point falls very close to the curve given by Dickson and Konopinski<sup>8</sup> for even atomic nuclei with one excess neutron. The predicted half-life for Ti<sup>45</sup> was five hours.

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<sup>&</sup>lt;sup>7</sup> H. Kopfermann and E. Rasmussen, Zeits. f. Physik **92**, 82 (1934).

<sup>&</sup>lt;sup>8</sup>G. R. Dickson and E. J. Konopinski, Phys. Rev. 58, 949 (1940).