The Radioactivity Induced in Silicon and Phosphorus by Deuteron Bombardment

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Silicon and phosphorus have been bombarded by deuterons and the resulting radioactive substances have been studied.

The two reactions,

$$Si^{30}+D^2=Si^{31}+H^1,$$
$$P^{31}+D^2=P^{32}+H^1,$$

were found to occur. The active substances were identified by the usual chemical tests. The half-life of Si^{31} was found to be 170 ± 10 minutes and that of P^{32}, 14.5 ± 0.3 days. Both substances were found to emit no gamma-rays and advantage was taken of this fact to study the absorption curves of the beta-rays with some care. The absorption curves showed very definite end points from which the maximum energy of the beta-ray spectra was calculated from Feather's empirical formula. The calculated energies were 1.50 ± 0.01 Mev and 1.59 ± 0.03 Mev for Si^{31} and P^{32}, respectively. These values are in satisfactory agreement with the latest direct measurements. The errors in absorption measurements in the presence of gamma-rays are estimated from the shape of the absorption curves.

THE ACTIVATION OF PHOSPHORUS

FROM a consideration of the types of reactions which have been studied heretofore, one may write only two by which radioactive products might be induced in phosphorus under deuteron bombardment:

$$P^{31}+D^2=P^{32}+H^1, \quad (1)$$

$$P^{31}+D^2=P^{30}+H^3. \quad (2)$$

All other reasonable reactions which might be written would lead to stable end products. Reaction (2) appears highly improbable, but it may be easily investigated since the period of P^{30} is known to be about 3 minutes.[2] A target of red phosphorus was bombarded for about five minutes with 3 Mev deuterons and the activity followed for about an hour. The decay curve became flat in about thirty minutes showing that an activity of relatively long period was present. On examining the earlier parts of the decay curve, weak activities with periods of about one minute and ten minutes were found. These are undoubtedly to be attributed to contamination of the target by oxygen and by carbon which give rise to F^{17} (half-life 1.16) and N^{13} (half-life 10.4), respectively. Since no trace was found of any half-life in the neighborhood of 3 minutes, it may be stated definitely that reaction (2) does not occur.

The long period which was found in this experiment is undoubtedly to be attributed to P^{32} formed by reaction (1). This substance has already been prepared in three ways:

$$Cl^{35}+n^1=P^{32}+He^4.$$
$$S^{32}+n^1=P^{32}+H^1,$$
$$Si^{29}+He^4=P^{32}+n^1.$$

The first two reactions were discovered by Fermi[3] and his collaborators while the third has been reported recently by Fallenbrach.[4] The half-life was found to be about two weeks.

In order to check the identity of the active substance obtained in these experiments, the target of red phosphorus was bombarded for an hour and the decay of the activity was followed for several days. As soon as it was apparent that the half-life of the substance was about the same as that reported for P^{32}, the sample was analyzed chemically. The red phosphorus was burned at the bottom of a test tube so that the oxide was condensed on the walls near the mouth of the tube. The deposit was dissolved in water and boiled with nitric acid in order to oxidize any

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[2] Curie and Joliot, Comptes rendus **198**, 254 (1934).
[3] Fermi, Amaldi, D'Agostino, Rassetti, and Segrè, Proc. Roy. Soc. **A146**, 483 (1934).
[4] Fallenbrach, Naturwiss. **23**, 288 (1935).

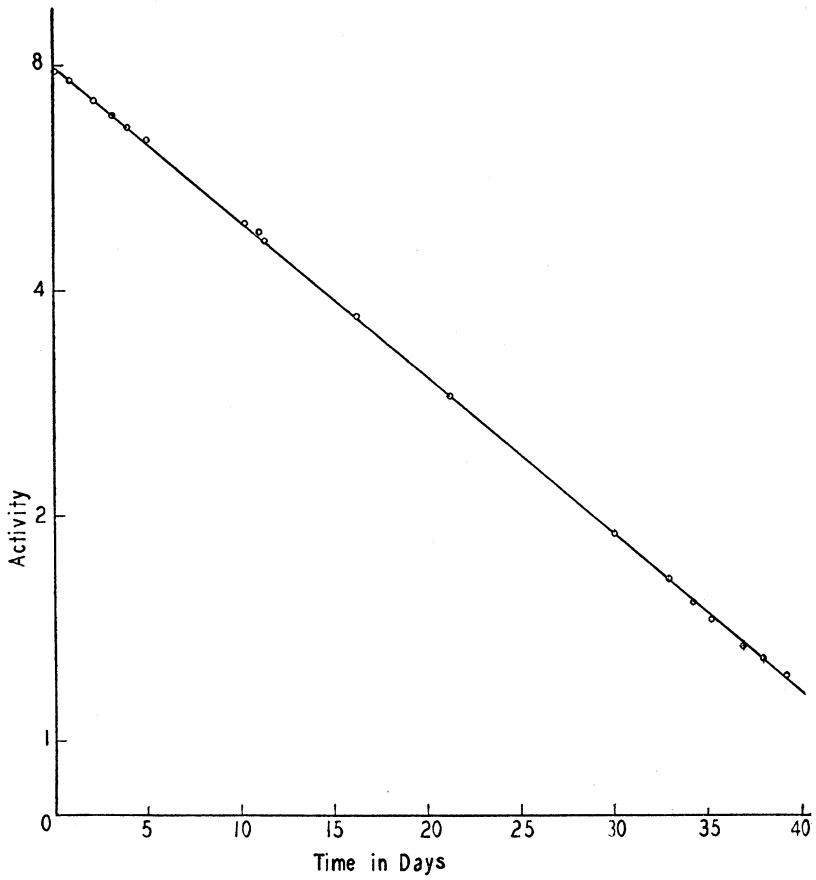

FIG. 1. A logarithmic plot of the decay of P^{32}.

material for which combustion was incomplete. Phosphates were then precipitated with magnesia mixture in the presence of small amounts of sulphates and chlorides. The precipitated $Mg(NH_4)(PO_4)$ showed a large activity. In order to show that the activity was not due to magnesium, aluminum or silicon, the precipitate was dissolved in dilute nitric acid and sodium silicate and aluminum chloride solutions were added. Silicon was then separated by evaporating the acid solution to dryness and filtering out the precipitated silicic acid. This precipitate showed no activity. The phosphates were then separated from aluminum and magnesium by precipitating $(NH_4)_3PO_4 \cdot 13MoO_3$ from dilute acid solution. This precipitate was found to contain all the activity showing that the active substance was an isotope of phosphorus.

In order to measure the half-life of P^{32}, an activated sample of phosphorus was sealed into the ionization chamber of an electroscope and the activity was followed for about three half-lives. The measurements were stopped when the activity was about twenty times the natural leak of the instrument in order that fluctuations in it would not introduce an appreciable error in the measurements. The decay curve so obtained is shown in Fig. 1. Each point on the curve is the average of ten measurements which differed from the mean by less than one percent. The half-life obtained from this curve is 14.5 ± 0.3 days.

The fact that no gamma-rays are given off by P^{32} (see later experiments) is sufficient evidence that the disintegration particles are electrons. However, magnetic experiments also show that the particles are negatively charged. The disintegration equation may then be written:

$$P^{32} = S^{32} + e^{-}.$$

The Activation of Silicon

Radioactive products might be expected to be induced in silicon by deuteron bombardment by the following reactions:

$$Si^{30}+D^2=Si^{31}+H^1, \quad (3)$$

$$Si^{30}+D^2=Al^{28}+He^4, \quad (4)$$

$$Si^{29}+D^2=P^{30}+n^1, \quad (5)$$

$$Si^{28}+D^2=P^{29}+n^1, \quad (6)$$

$$Si^{28}+D^2=Al^{26}+n^1, \quad (7)$$

$$Si^{28}+D^2=Si^{27}+H^3. \quad (8)$$

However, only the products of reactions (3) and (6) may be expected to be easily detectable. Si^{31} is known to have a half-life of about 3 hours[3] so that even though it arises from a rare isotope, the weak activity can be easily detected in the absence of other long periods. However, Al^{28} and P^{30} will be similarly weak and both of them have half-lives of about three minutes so that they will be detected only if other short periods are absent. Al^{26} has a very short half-life (about 7 seconds) and under the conditions of the experiment it probably cannot be detected. Reaction (8) seems very unlikely because the analogous reaction did not occur during the bombardment of phosphorus, and because the energy of the reaction is probably so negative that it could not be excited. The only remaining active substance is P^{29} which should be intense since it arises from the principal isotope of silicon. In the experiments, Si^{31} was easily detected after all of the shorter periods had disappeared. The only short period which was not obviously due to impurities was one of half-life 6–8 minutes and may have been due to P^{29}.

Strong sources of Si^{31} could be prepared by bombarding silicon targets for an hour or more and allowing about an hour for shorter periods to decay. The decay of the active substance could be followed for many half-lives, but owing to the presence of impurities of various substances, the half-lives obtained were not very reproducible. The best value for the half-life which has been obtained so far is 170 ± 10 minutes. This is to be compared with the value 150–180 minutes as found by Fermi[3] and his collaborators for the active substance from the reaction $P^{31}+n^1=Si^{31}+H^1$.

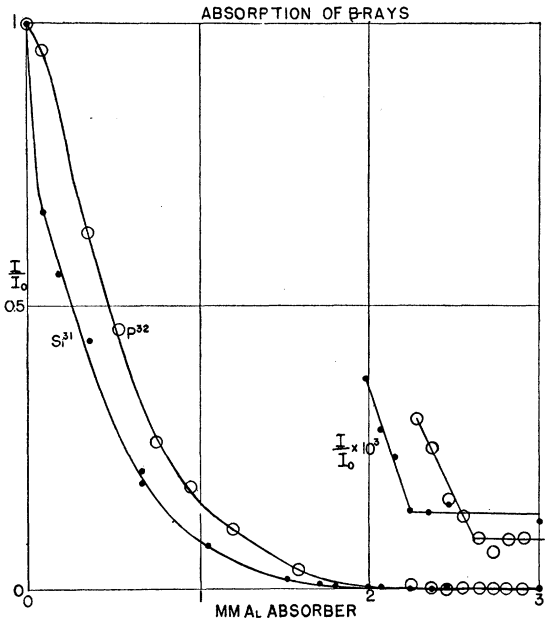

FIG. 2. The absorption of the electrons from Si^{31} and P^{32} in aluminum.

The chemical separation was carried out using the standard method for the analysis of silicon. The target was dissolved in concentrated potassium hydroxide, and silicic acid was precipitated in the presence of aluminum and phosphate ions, filtered in acid solution, and ignited in the usual manner. The activity of the silica which resulted was measured with an electroscope and found to be large. The silica was then treated with hydrofluoric and sulfuric acids to drive out all silicon as SiF_4. The residue was found to be completely inactive. This showed conclusively that the active substance of long life was an isotope of silicon; the filtrate was tested for active aluminium or phosphorus isotopes, but there were none present of long enough half-life to be detected at the end of this rather lengthy separation.

Cloud chamber experiments carried out by Kurie, Richardson, and Paxton showed that the disintegration particles are electrons and the absorption experiments which will be described confirm this result. The disintegration reaction is therefore:

$$Si^{31}=P^{31}+e^-.$$

The Absorption of Beta-Rays

Since both Si^{31} and P^{32} give off no gamma-rays, the absorption of the electrons may be measured accurately. Fig. 2 shows the absorption curves of the two substances in aluminum. From the large scale curves it will be seen that the ionization does not become truly constant until the intensity has been reduced to about one-thousandth of the original activity. This residual ionization, which is equal to about one-fifth of the natural leak of the electroscope, might be ascribed to weak gamma-rays, but experiments by McMillan[5] have shown that they are probably brehmstrahlen or x-rays due to the bombardment of the target material by β-rays. The end points of the absorption curves are very well marked and give maximum ranges in aluminum of 2.67 ± 0.04 mm and 2.51 ± 0.01 mm for P^{32} and Si^{31}, respectively. According to the empirical formula of Feather[6] these values correspond to energies of 1.59 ± 0.05 and 1.50 ± 0.01 Mev, respectively, while use of the range-energy relations of electrons leads to values slightly higher. These values are considerably lower than those obtained from the extrapolation of cloud track data according to the Uhlenbeck-Konopinski theory.[7]

It was thought that this discrepancy might be due to a high order of contact between the absorption curves and the axis as is predicted by the Uhlenbeck-Konopinski theory. To investigate this possibility, the absorption experiment was performed with two samples of Si^{31} where one sample was forty times as strong as the other. The result on the weaker sample gave a maximum energy of 1.44 ± 0.04 Mev as compared with the value for the intense sample previously given. The result showed a very slight increase in the maximum energy which was, however, nearly within the experimental error. It may be concluded, then, that the measured end point of the absorption curve is very close to the true end point. A careful determination of the maximum energy of the beta-rays from P^{32} has been made by Lyman[8] who finds a value of 1.7 Mev. The most recent result on the maximum energy of the beta-rays from Si^{31} has been made by Paxton[9] using cloud chamber data. He obtained a value of 1.65 Mev from the observed maximum energy of the energy distribution curves. The agreement between these two values and the results of the absorption experiments is very satisfactory considering the very different experimental conditions, and the fact that an empirical equation was used to obtain the energies of the electrons from their ranges.

It may be concluded from the above that reliable measurements on the maximum energy of beta-rays may be obtained by absorption measurements in the absence of gamma-rays. However, most artificially radioactive elements give off intense gamma-radiation, so that it is interesting to estimate what errors are introduced by the gamma-rays. If one gamma-ray quantum were given off per disintegration of P^{32}, the ionization due to the gamma would have been about one-fortieth of that due to the beta-rays. If one adds such a background to a logarithmic plot of the absorption of the beta-rays, it is found that, within the error of the instrument, the absorption curve meets the background at about 1.3 Mev instead of 1.6 Mev as found in the absence of gamma-rays.

In conclusion, I wish to express my gratitude to Professor Lawrence for the opportunity of using the cyclotron. I am also indebted to my colleagues at the Radiation Laboratory for both helpful discussion and practical assistance. The financial support of the Chemical Foundation and the Research Corporation is gratefully acknowledged.

[5] McMillan, Phys. Rev. **47**, 801 (1935).
[6] Feather, Phys. Rev. **35**, 1559 (1930).
[7] Kurie, Richardson and Paxton, Phys. Rev. **49**, 368 (1936).
[8] Lyman, Phys. Rev. **51**, 1 (1937).
[9] Paxton, Phys. Rev. **51**, 170 (1937).