# Radioactivity Induced in Sulphur

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Under bombardment of 4–6 Mev deuterons, sulphur becomes radioactive with the emission of both positive and negative electrons with 3 periods $3\pm0.1$ *m*, $33\pm1$ *m* and $14\pm0.3$ *d*. These decay periods were found to have their origin in $P^{30}$, $Cl^{34}$ (both positron emitters) and $P^{32}$ (negative electron emitter). The energy distribution of positrons from Cl was studied by a cloud chamber. When bombarded by neutrons two radioactive substances were produced which emit negative electrons. The half-value periods were found to be $2.6\pm0.2$ *h* and 14 *d*, which are characteristic of $Si^{31}$ and $P^{32}$.

## INTRODUCTION

SINCE the discovery by Curie-Joliot of artificial radioactivity, many radioactive isotopes have been found produced by bombardment with protons, deuterons, $\alpha$-particles and neutrons. Many light elements have been investigated in this laboratory by the use of the deuteron beam from the cyclotron. The work described in this paper is an investigation of radioactivity induced in sulphur by both deuteron and neutron bombardment.

## APPARATUS

The source of deuterons was the large cyclotron[1] in the Radiation Laboratory. At the time of the bombardment it was adjusted to furnish deuterons of energy of 4–6 Mev.

The targets were, in all cases except the first two exposures, sulphur fused on Pb plates and covered with two sheets of Pt foils, 4 mm in air equivalence, to avoid contaminations. In the first two cases, sulphur was fused on Al plates and covered with Al foils. The bombardments were made in vacuum on a special target holder with a cooling device to prevent the sample from evaporating.

All the measurements of activity were carried out on a Lauritsen type quartz fiber electroscope with an Al window and absorption measurements were made by placing Al sheets between the source and the window. A Wilson chamber[2] was used to investigate the nature of the ionizing particles from the irradiated samples and also for the measurement of the energy distribution of the positrons from radiochlorine.

[1] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608 (1934).

[2] I am indebted to Dr. Kurie for the use of the cloud chamber.

## I. ACTIVATION BY DEUTERONS

### Decay periods

A thick sulphur target on an Al plate was bombarded in vacuum at first for about 2 hours by an ion beam of 4 Mev in energy. At the end of the exposure the target was placed at the window of the electroscope. The measured activity as a function of time was plotted on a logarithmic scale and analysis of this curve shows the existence of two radioactive substances. The half-periods were observed to be $33\pm1$ *m* and $14\pm0.3$ *d*, and also there were traces of 2.6 *h* and 14 *h* activity. The ratio of the initial intensities of these substances was estimated roughly to be 360 : 1.6 : 0.3 : 0.2, respectively. Repeating the same exposure, the same type of decay curve was reproduced.

To find out whether there is a shorter period activity, a sulphur target on Pb covered with thin Pt foil, 0.0001″ in thickness, was bombarded for 10 minutes. After due allowance for the 33 *m* activity, a half-life of $3\pm0.1$ *m* was found. A short exposure of 5 *m* was made afterward and the same result was reproduced.

To investigate the nature of the ionizing particles, they were observed visually in a Wilson chamber which was placed in a magnetic field of a few hundred gauss. Both positrons and negative electrons were emitted from the target.

### The active substances

Ordinary sulphur contains three isotopes, $S^{32}$, $S^{33}$ and $S^{34}$, the relative abundance of which is reported to be 96 : 1 : 3. If sulphur combines

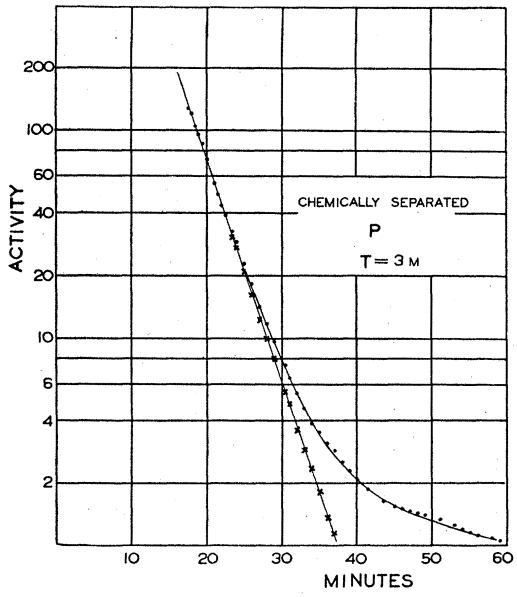


FIG. 1*A*. Logarithmic plot of the activity of the chemically separated $P^{30}$. Dots show the measured curve. *x*'s show the corrected curve for a small amount of contamination of Cl of about 1 percent in intensity.

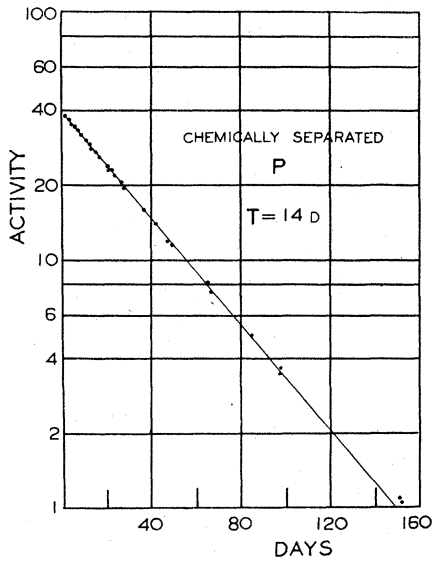


FIG. 1*B*. Logarithmic plot of the activity of the chemically separated $P^{32}$.

with deuterons, $Cl^{34}$, $Cl^{35}$, $Cl^{36}$ are expected to result with the capture of deuterons (this is supposed to be very improbable); $P^{30}$, $P^{31}$, $P^{32}$ with the emission of $\alpha$-particles; $Cl^{33}$, $Cl^{34}$, $Cl^{35}$ with the emission of neutrons; and $S^{33}$, $S^{34}$, $S^{35}$ with the emission of protons.

The identity of $P^{30}$ and $P^{32}$ is apparent since the periods characteristic of these substances are known with good accuracy. As for $Cl^{34}$, $Cl^{36}$ half-value periods of $40\pm5$ *m*[3] is given for $Cl^{34}$ and 50[4] to 35 minutes for $Cl^{36}$ (probably 37 *m*[5]). The accuracy of these measurements is not good enough to identify the 33 minutes activity.

Chemical tests were also made to check their identity. The sulphur sample was dissolved in $CS_2$, and mixed with dilute $HNO_3$, in which a small quantity of red phosphorus and a drop of dilute HCl was added. Then the solution was heated for a short time until the phosphorus was completely dissolved. After filtration of the water solution, Cl ions were precipitated with $AgNO_3$; then $PO_4$ ions were precipitated with magnesia mixture.

The logarithmic plot of the decay curves of these chemically separated samples evidenced excellent straight lines as shown in Fig. 1; consequently half-lives of $P^{30}$ (by short exposure), $P^{32}$ and Cl (by long exposure) were determined with sufficient accuracy. It is therefore obvious that the 3 *m* activity is due to $P^{30}$, 14 *d* to $P^{32}$ and 33 *m* to Cl.

Examining the chemically separated Cl in the Wilson chamber it was found that the sample emitted chiefly positrons and a small number of negative electrons. The latter can be explained as Compton recoil electrons of annihilation radiation of positrons and also a small amount of contamination of $P^{32}$. A study of the periodic table reveals that 33 *m* activity is probably due to $Cl^{34}$ or perhaps $Cl^{33}$, because the intensity of this Cl seems to be too strong to explain by the following process, if the fact is taken into account that the relative abundance of $S^{33}$ is only 1 percent: $S^{33}+D^2=Cl^{34}+n$.

Thus the reactions are supposed to be

$$S^{32}+D^2=P^{30}+He^4 \qquad P^{30}\rightarrow Si^{30}+e^+$$
$$S^{33}+D^2=Cl^{34}+n \quad (S^{32}+D^2=Cl^{33}+n^1 \quad S^{32}+D^2=Cl^{34})$$
$$Cl^{34}\rightarrow S^{34}+e^+$$
$$S^{34}+D^2=P^{32}+He^4 \qquad P^{32}\rightarrow S^{34}+e^-.$$

[3] O. R. Frisch, Nature **22**, 434 (1934).
[4] A. T. Alichanow, A. J. Alichanian and B. S. Dzelepow, Nature **135**, 393 (1935); E. Fermi and others, Proc. Roy. Soc. **A146**, 483 (1934); F. N. D. Kurie, J. R. Richardson and H. C. Paxton, Phys. Rev. **47**, 796 (1935).
[5] A. R. Olson, W. F. Libby, F. A. Long, R. S. Halford, J. Am. Chem. Soc. **58**, 1313 (1936).

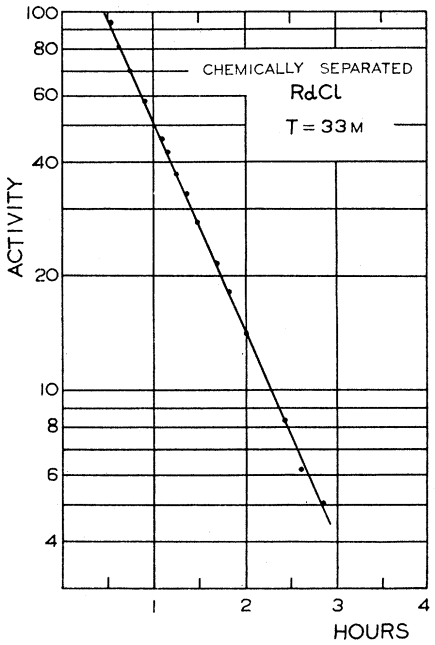


FIG. 1C. Logarithmic plot of the activity of the chemically separated radiochlorine.

$P^{32}$ can also be expected to result from a neutron bombardment on sulphur and there is a probability that $P^{32}$ might be created by neutrons from D+D. An experiment to test this point was made. Two similar samples of sulphur were put one upon another on the target holder, and the upper target was bombarded by deuterons. As these targets were rather thin, intensities on both targets of the neutrons created on the upper target by D+D were supposed to be the same order of magnitude. After bombardment the intensities of the activity of the lower target was found to be less than 1/200 of the upper target. This definitely demonstrated that $P^{32}$ on the upper target was chiefly created by deuteron impacts.

**The radioactive radiations**

The beta- and gamma-rays from $P^{30}$ and $P^{32}$ have been studied in detail by many others and there is no need to reiterate it here. However, Frisch is the only person who has succeeded in making $Cl^{34}$. He created it by bombardment of $P^{31}$ by $\alpha$-particles, consequently the intensity must be very weak and the accuracy of the measurement is rather poor. In the present experiment radioactive chlorine was made by deuterons with strong intensity, the initial intensity of 2 hours' exposure being 300 divisions per second, that is $3\times10^4$ times the natural leak. Some detailed investigation of the radiations from radioactive chlorine was made.

*Gamma-rays.* Rather weak gamma-rays were found, the absorption coefficient of which was determined with lead sheets. A value of 5 mm of lead for the half-value thickness was obtained. Although the geometrical conditions were not satisfactory, this value probably corresponds to an energy of about 0.5 Mev. This is in agreement with the energy of the gamma-rays which is expected when positrons from Cl are annihilated.

*Beta-rays.* The energy distribution of $\beta$-rays from radioactive chlorine was studied by the use of the cloud chamber with a magnetic field of about 400 gauss. The source was chemically separated from the irradiated sulphur in half an hour after exposure and then mounted inside of the chamber, covered with a thin Al foil to protect the chamber from contamination. This source was rather thick owing to the addition of too much chlorine in the course of chemical separation and one could not get rid of some irregularity in absorption; still the result shows a typical positron distribution and the Konopinski-Uhlenbeck[6] plot gives a fairly good straight line as shown in Fig. 3.

The upper limit is estimated to be 3 Mev. The point corresponding to 3 Mev and 33 *m* placed on Sargent's[7] curves of beta-ray periods and energies lies right on the second group line.

## II. ACTIVATION BY NEUTRONS

A block of sulphur (20×15×3 mm) was put about 5 cm from the Be target of the cyclotron and irradiated for about 2 hours by neutrons. After irradiation the activity was measured as before. The decay curve was analyzed in two exponential curves showing the existence of two radioactive substances, the decay periods of which are 2.6 hours and 14 days.

These periods are characteristic of $Si^{31}$ and $P^{32}$ which are expected by the following reactions.

[6] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 107 (1935); F. N. D. Kurie, J. R. Richardson and H. C. Paxton, Phys. Rev. **49**, 368 (1936).
[7] B. W. Sargent, Proc. Roy. Soc. **139**, 659 (1933).

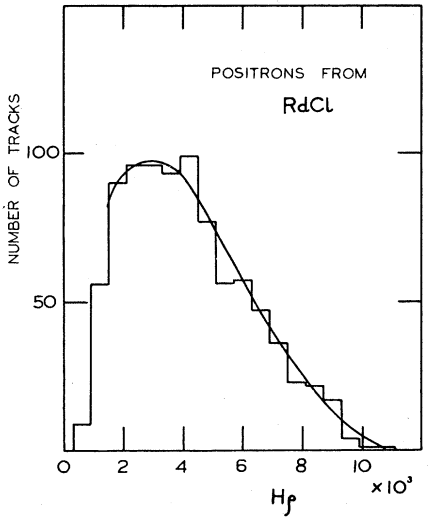


FIG. 2. Distribution histogram for the positrons emitted from chemically separated radiochlorine.

$$S^{34}+n=Si^{31}+He^{4},$$
$$S^{32}+n=P^{32}+H^{1}.$$

Rough chemical tests were made by dissolving activated samples with $CS_2$. A small quantity of Si powder and red phosphorus was added, as a mixture in dilute $HNO_3$. Then it was heated slightly till the powder of phosphorus was dissolved completely by $HNO_3$. After filtration of $HNO_3$ solution, phosphorus was precipitated by the magnesia mixture. The $CS_2$ solution was also filtered afterward and the Si powder was obtained as a residue.

By examining these samples it was found that the 14-day activity was due to a phosphorus isotope and the 2.6 hour activity to an Si isotope.

As for the $CS_2$ solution, it was dried to powder of sulphur. This was then oxidized by sodium chlorate and $HNO_3$ to $H_2SO_4$. The latter was precipitated with $BaCl_2$ as $BaSO_4$ in HCl solution. But this sample showed practically no activity.

### A search for $S^{35}$

According to the consideration of reactions with S, a radioactive sulphur $S^{35}$, which is said to have a long half-life of about 80 days,[8] is expected to result from both deuteron and neutron bombardment.

A search for this isotope was made by making a careful chemical separation of sulphur as before. The first two or three chemically separated samples showed some small activity, but by repeating the process more carefully the activity was found to be much smaller than before and by measurement of the half-life, this activity was found to be due to the contamination of $P^{32}$.

The result shows that the deuteron bombardment for 3 hours with 13 microamperes beam on the target, cannot make $S^{35}$ of intensity of more than 1/1000 of $P^{32}$.

As for neutron bombardment, the longest exposure was made on one sample which was put about 10 cm from the Be target of the cyclotron for about 20 days completely covered with paraffin. During these days the cyclotron ran very steadily with about 10–15 microamperes of 5.5 Mev deuterons and nearly every day neutron bombardment on mice and wheat seeds, etc., was continued. Yet the activity of the chemically separated sulphur from this sample was about the same order of magnitude of the natural leak and even this activity was also found afterward to be due chiefly to $P^{32}$ contamination.

In case of this thick target, it is quite difficult to estimate the lower limit, but it seems the cross

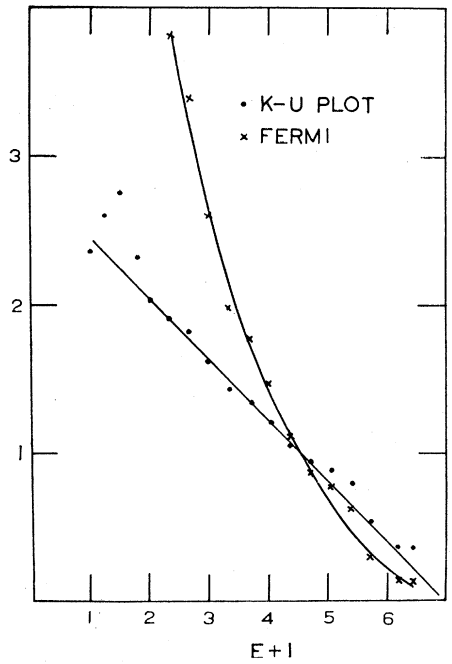


FIG. 3. $K-U$ plot for radiochlorine. This plot extrapolates to an upper limit at $E+I=6.9$, about 3 Mev.

[8] E. B. Andersen, Zeits. f. physik. Chemie **B32**, 237 (1936).

section for neutron capture by sulphur must be 1/100 of that for excitation of $P^{32}$ although the samples was surrounded by paraffin.

ACKNOWLEDGMENT

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# A Positive Energy Wave Packet Solution of the Dirac Equation

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A wave packet solution of the Dirac equation is obtained by superposition of positive energy solutions only. The solution involves an arbitrary parameter, and it is shown that this can be chosen so that all three dimensions of the packet are less than $\hbar/mc$. The possibility of constructing such a packet had been doubted. It is also shown that in the limit $\Delta x=0$, $\Delta x\Delta p_x=1.49\hbar$. It is not known whether this is the smallest value for the uncertainty product, subject to the condition that only positive energy solutions are used in the construction of the packet, or whether some other solution might yield a still smaller value.

A QUESTION of some technical interest is the following: can a wave packet be constructed by superposing only positive energy solutions of the Dirac equation for the free electron, such that all its dimensions are smaller than $\hbar/mc$? Apparently no conclusive answer to this question has been published.[1] In this note it will be shown that the omission of the negative energy solutions does not impose a lower limit on the size of the wave packet. This will be done by constructing a particular wave packet whose dimension at $t=0$ is arbitrary, and using only positive energy solutions in its construction.

Because of the relativistic invariance of the theory, it is sufficient to consider only packets whose centroids are at rest in the system of coordinates chosen, It is known that in general, that packet for which $\Delta x\Delta p_x$ is smallest is obtained by using a Maxwellian distribution of momenta. In a preliminary investigation, it was found very difficult to discover the analog of this theorem when the positive energy condition is imposed. It was therefore decided to use a distribution such that the Maxwellian is approached for large packets. A further consideration was that all necessary integrals should be calculable without dubious approximations.

In the following, $\hbar/mc$ will be used as unit of length, and $\hbar/mc^2$ as unit of time. Then the Dirac equation can be transformed to the Schrödinger-Gordon equation by the substitution

$$\psi=i[(\partial F/\partial t)+\boldsymbol{\alpha}\cdot\nabla F]-\alpha_m F. \tag{1}$$

In order that $\psi$ satisfy the Dirac equation, the "spinor potential" $F$ must satisfy

$$(\partial^2 F/\partial t^2)=\nabla^2 F-F. \tag{2}$$

Wave packet solutions of this equation are easily written down in the form

$$F_\sigma=\int G_\sigma(\mathbf{p})\exp[i(\mathbf{p}\cdot\mathbf{x}\pm wt)]d\mathbf{p}, \tag{3}$$

where $\sigma$ is the spin variable, and

$$w=(1+p^2)^{\frac{1}{2}}>0. \tag{4}$$

If the negative sign of $w$ is chosen in Eq. (3), then $\psi$ will involve only positive energy solutions.

For the present purpose, it is sufficient to take

$$G_1=A\exp(-\beta w),\qquad G_2=G_3=G_4=0, \tag{5}$$

[1] It is known that several investigators have arrived at conflicting conclusions regarding this question. The only definite expression of opinion that has been published seems to be that of L. de Broglie, *L'Électron Magnétique*, p. 286.