to a very small value as n becomes large. Table VI shows the values of these frequencies in the different electronic states. The values of ν_1 and ν_5 in the ground state are assumed to be 1623 cm^{-1} and 1342 cm^{-1}.[11]

TABLE VI. *Values of ν_1 and ν_5 frequencies.*

n	2	3	4	5	6
ν_1	1340	800	300	230	200
ν_5	480	150	—	—	—

A large number of weak diffuse bands were obtained which have not been classified. Some of them undoubtedly correspond to other electronic series.

THE ABSORPTION OF ETHANE

At very low pressures (0.01 mm) the absorption of ethane consists of extremely diffuse bands starting at 1350A and going down below 1000A. The bands are far too diffuse for any analysis to be attempted. In this respect they resemble methane.[12] The low pressure bands in methane start at 1300A; at somewhat higher pressures they extend up to 1450A. Those of ethane behave in the same way and move to a limiting value of around 1500A with increase in pressure. On account of the extreme diffuseness of the bands in ethane it is assumed that as in methane all the upper electronic states are unstable.

In conclusion the author wishes to express his gratitude to Dr. G. H. Dieke for his generous help and numerous suggestions in connection with both the experimental and theoretical aspects of this work.

[11] Mecke, Zeits. f. physik. Chemie **B17**, 16A (1932).

[12] A. B. F. Duncan, J. Chem. Phys. **2**, 851 (1934).

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Artificial Radioactivity Produced by the Deuteron Bombardment of Nitrogen[1]

EDWIN McMILLAN AND M. STANLEY LIVINGSTON,* *Radiation Laboratory, Department of Physics, University of California*

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This paper describes the work leading to the discovery and investigation of radio-oxygen, and also includes some incidental results on other nuclear disintegrations. It is shown that nitrogen bombarded with deuterons gives rise to a radioactive substance which emits positive electrons of maximum energy 1.2 mv, has a half-life of 126±5 sec., and is found by chemical analysis to be an isotope of oxygen. The nuclear cross section for the activation of nitrogen at 2 mv deuteron energy is 6×10^{-27} cm^2; that for the activation of carbon is three times as great. The neutrons expected to accompany the formation of radio-oxygen were found to be present. These results were obtained using gaseous nitrogen as a target; in some of the experiments use was made of the fact that the active product can be deposited on a metal surface by recoil.

EXPERIMENTAL ARRANGEMENT

DEUTERONS were accelerated to a kinetic energy of about 2 mv in an apparatus described elsewhere.[2] A brass tube was set into the side of this apparatus in such a position as to intercept the deuteron beam, which passed into the atmosphere through a vacuum-tight window[3] as shown in Fig. 1. The target to be

* Now at Cornell University.

[1] A preliminary report of this work was given by M. S. Livingston and E. McMillan, Phys. Rev. **46**, 437 (1934).

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

[3] The window used for admitting the deuteron beam was so satisfactory that it seems worth while to describe it in some detail. It consisted of a sheet of 0.0001″ thick aluminum foil (procured from the American Platinum Works) mounted with wax on a brass plate, covered as closely as possible with one mm holes over an area of 1×1.5 cm. A wax to be used for this purpose must have a high melting point (the window becomes quite warm under the deuteron bombardment), it must flow freely when melted, and it must stick firmly to a clean metal surface when cold. These requirements are satisfied by rosin, with just enough Venice turpentine added to make it sticky, as determined by trial. The window was mounted by heating the supporting grid to a temperature above the melting point of the wax, covering it with a thin coat of wax and blowing off the excess, and then putting on the foil, touching it at one edge first and

activated was placed opposite the window. The space containing the target could be evacuated or filled with various gases during the bombardment.

For studying the activity produced in gases by deuteron bombardment, a plug shown as a dotted line in Fig. 1 was waxed into the tube, leaving a space in which the gases could be bombarded. Fig. 2 shows the gas-handling arrangement used. After evacuating the system, the activating chamber was filled with the gas to one atmosphere pressure through the line G.

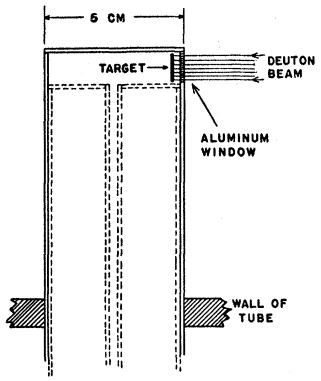

FIG. 1. Arrangement for bombarding targets outside the vacuum. A plug which may be put in to enclose a space for bombarding gases is shown by dotted lines.

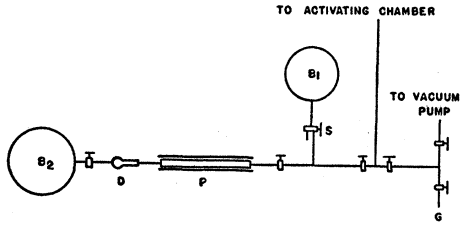

FIG. 2. Gas-handling system. G, line for introducing gases into the activating chamber. B_1 and B_2, bulbs for collecting activated gases before and after passage through the analyzing train consisting of the platinized asbestos tube P and $CaCl_2$ drying tube D.

allowing the surface tension of the wax to pull it into place. A small leak in one hole of the window can be stopped by filling the hole with a drop of wax, applied with a hot pointed iron. A window constructed in this way has a large open area, a small stopping power (4 mm air equivalent) and can be exposed to several microamperes of ion current for long periods with no apparent damage. This seems remarkable at first sight since the wax on the back of the grid rapidly becomes carbonized and the beam in air will char paper in a few seconds, but it appears reasonable when one considers the small amount of energy lost by a high speed ion in going through the foil.

When the bombardment with deuterons was completed, the activated sample was allowed to flow into the bulb B_1, whose volume was sufficiently large to collect over 90 percent of the sample. This bulb was then removed and placed beside the electroscope, which was discharged by the emitted gamma-rays. The part of the system to the left of B_1 was used for the chemical analysis, and will be described later in the paper.

IONIZATION CHAMBER CALIBRATION

The ionization chamber used for studying the radioactivity consisted of a cylinder of dimensions 7×7 cm containing a sensitive (2×10^6 ion pairs/div.) Lauritsen type quartz fiber electroscope and provided on one side with a large window covered with 0.001 inch thick aluminum foil. From the geometry of the arrangement and the known ionization per centimeter of beta-particles one can compute roughly that, with an active target next to the window, a leak of one division corresponds to a total of 2×10^4 beta-disintegrations in the target.

For evaluating the measurements made with the gamma-rays from a gas sample contained in a bulb, a calibration of the following sort was made: An activated carbon target (which is known to give no strong gamma-rays except the "annihilation radiation" associated with the positive electrons emitted as beta-particles) was placed next to the window of the electroscope and the ionization observed. The same target was then enclosed in a box thick enough to stop all beta-particles and placed in the position of the center of the gas bulb and the ionization again observed. The ratio of the observed ionizations was 100. It was found also that the ionization produced by the target when enclosed in a box next to the window was 1/35 that produced by the bare target in the same position. This number gives a criterion for deciding whether a positron-emitter gives any appreciable gamma-rays other than the annihilation radiation.

RADIOACTIVITY PRODUCED ON SOLID TARGETS

Carbon was the first substance to be activated after the installation of the arrangement of Fig. 1. A beam of about $\frac{1}{2}\mu$A of 2 mv deuterons emerging from the window was used, and the carbon

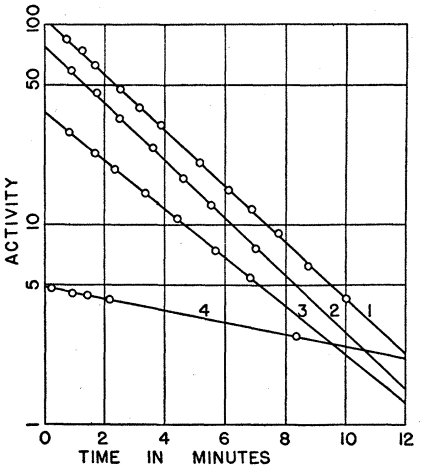

FIG. 3. Logarithmic decay curves of the activities of: 1. Platinum bombarded in air. 2. A bulb containing activated air. 3. Platinum bombarded in vacuum. 4. Platinum bombarded in CO_2. The absolute values of the activities are not represented in their correct ratios.

target was found to give, as expected,[4] a very large activity decaying with a half-life of 10′ 23″±5″. The rate of leak of the electroscope with the target next to the window of the ionization chamber, corrected to what it would be immediately after a very long exposure to $\frac{1}{2}\mu$A of deuterons,[5] was 600 divisions per second, corresponding to an activation probability of 3.6 per million deuterons striking the target. An aluminum target gave an activity with a half-life of 156±5 sec. and an intensity of 50 div./sec.[6]

After this an investigation of possible contamination effects in this type of experiment was undertaken. A piece of platinum sheet was cleaned by holding it at white heat for several minutes in an oxygen flame, and it was then bombarded with deuterons. It showed an activity of 5 div./sec., decaying with a half-life of 126 ±5 sec. Curve 1 of Fig. 3 is a logarithmic plot of the decay of this effect showing by its linearity that there was only one decay period present and that there was no appreciable carbon contamination. Clean copper and oxidized copper gave identical effects. This made it seem probable that the observed activity came from the gas surrounding the target, and therefore the effects produced on a platinum target when bombarded in various gases were examined. Displacing the air with O_2, H_2 and A reduced the effect to nearly zero; N_2 gave the same effect as air, and CO_2 gave an effect of the same magnitude (5 div./sec.) but decaying with the characteristic carbon period of 10.4 min. Curve 4, Fig. 3, shows the decay of the activity found on platinum bombarded in CO_2.

INTERPRETATION OF THE EFFECT OBSERVED ON PLATINUM

The foregoing results make it obvious that the activity found on platinum when bombarded in air arises from nitrogen and is deposited on the target surface in some way. The existence of convection currents due to the heating of the target and window makes it unlikely that activated atoms in the gas would remain near the target long enough to diffuse to its surface and be adsorbed. It was also found that the active substance was firmly attached to the platinum, being unaffected by hot HNO_3 and only half removed by heating the target to white heat for 10 seconds.

These observations suggested that the activated nuclei were driven into the target surface from the gas by the recoil momentum imparted by the incident deuterons. The following experiment established this mechanism with certainty. A platinum target bombarded in vacuum showed an activity of 2.5 div./sec. decaying with the 156 second period of activated aluminum (curve 3, Fig. 3). This could come only from the aluminum window, and could be transferred onto the target only by recoil.

THE RADIATIONS FROM ACTIVATED NITROGEN

The beta-particles from a platinum target containing activated nitrogen were found by magnetic deflection to have a positive sign. Their absorption in aluminum is shown in Fig. 4, together with that of the beta-particles from activated carbon. From these curves one cannot make a precise estimate of the maximum ranges, but by extrapolating according to the known

[4] H. R. Crane and C. C. Lauritsen, Phys. Rev. **45**, 430 (1934); J. D. Cockcroft, C. W. Gilbert and E. T. S. Walton, Nature **133**, 328 (1934); M. C. Henderson, M. S. Livingston and E. O. Lawrence, Phys. Rev. **45**, 437 (1934).

[5] All rates of leak given in this paper are corrected in the same way.

[6] Further information about the aluminum activity is given by E. McMillan and E. O. Lawrence, Phys. Rev. **47**, 343 (1935).

shape of beta-absorption curves (and taking account of the gamma-ray background) one sees that they cannot be far different from 0.5 g/cm² for activated nitrogen and 0.4 g/cm² for activated carbon. The upper energy limits computed from these values by Feather's[7] formula are 1.2 and 1.0 mv, respectively. The value 1.0 mv for activated carbon is not in perfect agreement with Anderson and Neddermeyer's[8] value 1.2 mv obtained by measuring the curvature of cloud chamber tracks in a magnetic field; whether this discrepancy arises from an underestimate of the maximum range given by our absorption curve or from scattering of the beta-particles producing Anderson and Neddermeyer's tracks must await further experiment for a decision.

Our upper limit of 1.2 mv for activated nitrogen, together with the half-life of 126 sec., places the active substance very slightly above Sargent's[9] upper curve.

The gamma-rays from activated nitrogen produce the same ionization relative to that of the beta-particles as is observed in the case of carbon, which indicates that no intense gamma-ray is present other than the annihilation radiation associated with the emitted positrons (2 quanta of $\frac{1}{2}$mv radiation per beta-particle).

Activation of Gas Targets

Samples of gases activated, transferred to a bulb, and examined by means of their gamma-radiation, gave the following activities: Air, period 126 sec. and activity 0.5 div./sec. (decay shown by curve 2, Fig. 3); nitrogen, the same; CO_2, period 10.4 min. and activity 0.25 div./sec.[10] If one compares the value for CO_2 with the activity produced in a solid carbon target, using the conversion factor between beta- and gamma-ray measurements mentioned above, and remembering that the stopping power per carbon atom is 3.5 times as great in CO_2 as in C, it appears that

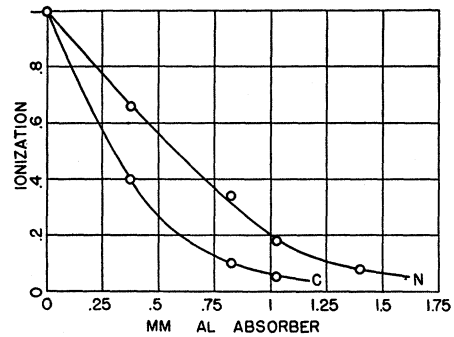

FIG. 4. Absorption in aluminum of the beta-particles from activated nitrogen and carbon.

only one-seventh of the activity produced in the gas passes into the bulb. This is undoubtedly explained by the fact that the active product (N^{13}) is left in the monatomic state after its formation; in this state it is chemically reactive and has a large chance of combining with the brass wall of the activating chamber before forming a stable gaseous compound. Because of this fact no good estimates of activation probabilities can be made from the gas-target experiments.

Chemical Identification of the Active Product from Nitrogen

The active substance produced from nitrogen must be an isotope of C, N or O if it is formed in any expected type of nuclear reaction. A very simple gas analysis was able to show that it is definitely oxygen. A sample of air was activated and allowed to flow into the bulb B_1 (Fig. 2). Then the activating chamber was filled to one atmosphere with hydrogen, which was also let into B_1. The resulting mixture was then passed by way of a platinized asbestos tube P and a $CaCl_2$ drying tube D into the previously evacuated bulb B_2 and flushed through with nitrogen admitted at S. The drying tube and the bulb B_2 were then removed and examined separately for activity. The result was that with the platinized asbestos tube heated to 500°C (being then a very efficient catalyst for the combination of oxygen and hydrogen to form water) the activity went quantitatively into the drying tube; with the platinized asbestos tube cold, it went equally quantitatively into B_2. This is what would be

[7] N. Feather, Phys. Rev. **35**, 1559 (1930).
[8] C. D. Anderson and S. H. Neddermeyer, Phys. Rev. **45**, 653 (1934).
[9] B. W. Sargent, Proc. Roy. Soc. **A139**, 659 (1933).
[10] A search was made also for possible activity produced in deuterium by deuteron bombardment. The activating chamber was filled with $\frac{1}{2}$ atmosphere of deuterium, and no activity was found other than a small effect of 0.01 div./sec. with the carbon period, probably coming from a trace of wax on the surface of the window.

expected of an active oxygen. The added hydrogen is more than enough to convert all the oxygen in the air sample into water; and the active oxygen, even if present as N_2O or NO_2 because of its chemical reactivity after formation, would be carried along with it. On the other hand, carbon (as CO or CO_2) would be reduced to free carbon or CO, and nitrogen or its oxides to N_2 (the formation of ammonia is very slight under these conditions), and none of these would go into the $CaCl_2$ tube.

Nuclear Reaction for the Formation of Radio-Oxygen

The reactions involved in the formation and disintegration of this active oxygen are presumably:

$$N^{14}+H^2\rightarrow O^{15}+n, \quad (1)$$

$$O^{15}\rightarrow N^{15}+e^+. \quad (2)$$

Since nothing is known about the energy of the emitted neutrons, a mass for O^{15} cannot be calculated. But the energy available in the total process (1)+(2) can be computed, taking $N^{14}=14.0074$, $H^2=2.0136$, $n=1.008$ and $N^{15}=15.0041$.[11] It is found in this way to be 7.3 mv.

Yield of Radio-Oxygen

As explained above, the magnitude of the gas-target effect does not give a trustworthy measure of the activation yield, but it suggests that this yield is of the same order as that for carbon. A better estimate can be made from the activity deposited by recoil. The mean forward momentum of the O^{15} nuclei immediately after formation is 15/16 that of the incident deuterons, the momentum imparted by the expelled neutrons being random in direction and therefore canceling out. The mean forward range of the recoil O^{15} nuclei activated by 2 mv deuterons is accordingly, to a first approximation at least, one mm of air. Nearly the same value is obtained for the N^{13} nuclei produced by the activation of carbon. This means that the activity deposited on platinum in N_2 or CO_2 comes effectively from a layer of the gas of thickness one mm air equivalent.

Since the equilibrium activity of a one mm layer of air when bombarded by $\frac{1}{2}\mu$A of 2 mv deuterons is thus found to be 5 div./sec. (10^5 disintegrations/sec.), the cross section for the activation of nitrogen at this deuteron energy is 6×10^{-27} cm^2. The corresponding cross section for carbon is three times as great, since the same recoil activity is found on platinum bombarded in CO_2, and the number of carbon atoms per unit stopping power in this substance is only one-third as great as the corresponding number of nitrogen atoms in air.

The total activity produced in nitrogen over the whole deuteron range cannot be calculated from the available data, but it can be estimated by assuming that the form of the activation function for nitrogen is about the same as that for carbon. On this assumption the total yields for nitrogen and carbon would be in the same ratio as their cross sections at the maximum energy; the total yield found in this way for 2 mv deuterons in N_2 is one activation per million deuterons. The actual value is certainly somewhat smaller, since the nitrogen activation function is expected to fall off more rapidly at low voltages than that for carbon.

These values of the cross sections and yields cannot be considered as precise, and may easily be wrong by a factor of two. Some justification of the calculations involving the recoil ranges is given by the fact that in the case of Al[6] it was possible to check the computed recoil range experimentally.

Attempt to Activate a Solid Target Containing Nitrogen

The yield values given above suggested that a target of a solid nitrogen compound could be activated to considerable intensity. Trying this with NH_4NO_3 and KNO_3 we found an activity of 8 divisions per second, scarcely larger than that deposited by recoil on platinum. This curious result may mean that oxygen can diffuse readily through a salt of this sort, so that the active substance "evaporates" rapidly from the target.

[11] From the disintegration data of E. O. Lawrence, E. McMillan and M. C. Henderson, Phys. Rev. **47**, 273 (1935).

The Neutrons from Nitrogen

Some evidence for the existence of the neutrons indicated by Eq. (1) has been given by earlier work in this laboratory.[12] A further investigation was made in connection with the present report. The neutron emission was observed by a Wynn-Williams ionization chamber (with a paraffin sheet in front) and linear amplifier, connected to a thyratron scale-of-eight counter. At first an attempt was made to measure the neutrons from gases contained in the activating chamber, placing the counter as close to it as possible (about 15 cm). The numbers of counts obtained with the chamber full of air, CO_2, H_2[2]($\frac{1}{2}$ atmos.),[13] and evacuated were the same within the constancy of the measurements and were one-sixth as large as the number of counts obtained with a beryllium target in the path of the beam in vacuum just inside the window. This was interpreted as showing that the neutron background (arising partly from contamination throughout the apparatus and partly from the wax and aluminum of the window) was larger than the effects from the gas targets at that distance. It was therefore necessary to place the counter closer to the bombarded gas to obtain significant results. This was done by sliding the counter into the brass tube containing the window, to within 3 cm of the deuteron beam.

With this arrangement counts were made with the bombarding space full of air, nitrogen and oxygen, and also with a clean sheet of copper stopping the beam next to the window. With a deuteron beam of 0.1μ A, the following counting rates were obtained: Copper sheet in place, 500 counts per minute; oxygen target, 500 counts per minute; nitrogen and air targets 700 counts per minute. The difference of 200 counts per minute is attributed to the neutrons from nitrogen; its reality was checked by taking many alternate counts with the copper sheet in and out.

A value for the total neutron yield from nitrogen bombarded by 2 mv deuterons can be obtained from this result if one assumes that one neutron is counted per thousand traversing the counter and notes that the fraction of the total solid angle subtended by the counter at the target is 1/50. The resulting value for the yield is 0.3 per million deuterons; the difference between this and the yield of radioactivity is certainly within the accuracy of these estimates.

Acknowledgments

The authors wish to express their sincere appreciation to Professor E. O. Lawrence for the opportunity of doing this work and to the Research Corporation and the Chemical Foundation for their helpful financial support.

[12] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 220 (1934).

[13] This observation of the neutrons from deuterium was made to see whether the enormously large yields reported by other investigators for lower energy deuterons are also produced at 2 mv; it shows that the yield in this case is at most one-sixth of that from beryllium, and is probably less. It must be noted that with the activating chamber filled with deuterium to $\frac{1}{2}$ atmosphere the deuterons use only one-tenth of their range in traversing the gas, since they are just able to cross the chamber when it is filled with air at atmospheric pressure.