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Transmutations of Aluminum by Deutons

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Aluminum bombarded by 2.2 mv deutons is found to emit protons, neutrons and alpha-particles, presumably accompanying reactions in which Al^{28} (radio-aluminum), Si^{28} and Mg^{25} are formed. The protons are the most abundant (about 8 per 10^7 deutons) and have a complex spectrum of ranges, in which groups with ranges 10, 21, 30 and 53 cm have been resolved. The maximum range of 62 cm corresponds to a reaction energy of 5.3 mv. The radioactive product, shown chemically to be an aluminum isotope, decays with a half-life of 156 ± 5 sec., emitting negative electrons and intense gamma-rays. The maximum range of the electrons is 0.95 g/cm^2 of Al, corresponding to an energy limit of 2 mv. The excitation curve for the radioactivity agrees with Gamow's theory of nuclear penetration. The alpha-particles have a range of 6.5 cm, giving a reaction energy of 6.6 mv for the formation of Mg^{25}. They are only 1/100 as abundant as the protons.

INTRODUCTION

IN earlier experiments in this laboratory it has been found that aluminum under deuton bombardment gives off alpha-particles,[1] protons and neutrons,[2] and becomes radioactive.[3] This paper describes a more detailed investigation of these reactions.

The apparatus used for producing a beam of high speed deutons is described elsewhere.[4] For studying the emission of particles from the target during bombardment, the target was placed in the vacuum in the path of the deuton beam and the disintegration particles were allowed to emerge through a mica window, as shown in Fig. 1. For studying the radioactivity, the beam was sent through a thin aluminum window (about 1 cm air equivalent stopping power) so that the target, placed opposite the window, could be bombarded outside the vacuum, and therefore could be removed from the apparatus immediately after exposure.

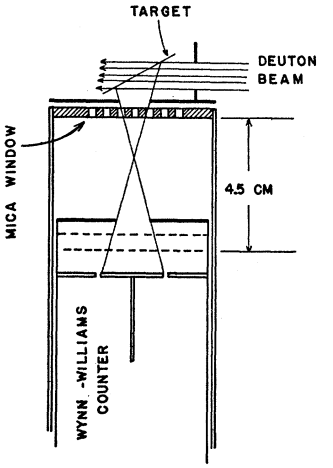

FIG. 1. Arrangement of target, screens and counter for bombarding in vacuum.

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[1] G. N. Lewis, M. S. Livingston and E. O. Lawrence, Phys. Rev. **44**, 55 (1933).

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

[3] M. C. Henderson, M. S. Livingston and E. O. Lawrence, Phys. Rev. **45**, 428 (1934).

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

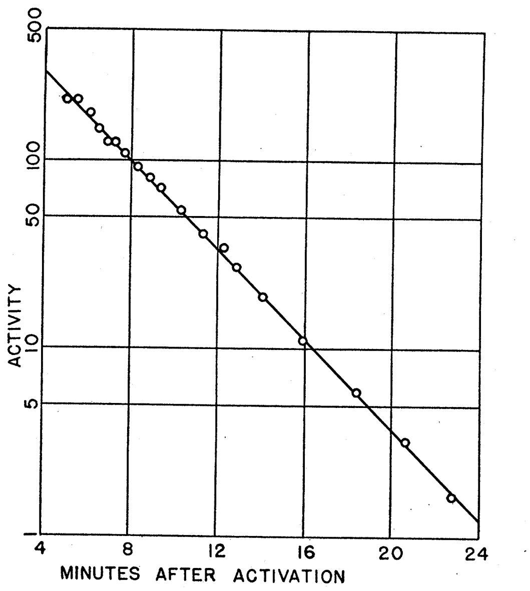

FIG. 2. Decay curve of Al radioactivity, with ordinates on a logarithmic scale.

RADIOACTIVITY

An aluminum target, exposed to ½ microampere of 1.9 mv deutons emerging from the window for a sufficient time to give saturation activity, was placed next to an ionization chamber containing a sensitive quartz fiber electroscope and provided with a thin aluminum window to admit beta-particles. The rate of discharge of the electroscope was followed over a sufficient time to allow the activity to decay by a factor of 120. A logarithmic plot of discharge rate against time (Fig. 2) was accurately linear, giving a half-life for the activity of 156±5 sec. The initial rate of discharge, extrapolated back to the time of removal from the deuton beam, was 50 divisions per second. One can compute roughly from the sensitivity (one division $=2\times10^6$ ion pairs), the geometry of the electroscope, and the ionization per centimeter of beta-particles, that this corresponds to a rate of activation of about 3 radioactive atoms per 10^7 deutons striking the target.

THE RADIOACTIVE RADIATIONS

The beta-particles were found by magnetic deflection to have a negative sign. Their absorption in aluminum was studied by placing foils between the target and the ionization chamber, with the results shown in Fig. 3. The curve obtained by Lawrence[5] for radio-sodium is given for comparison. As in the case of sodium, the aluminum beta-particles are accompanied by an intense gamma-radiation,[6] so that the ionization does not go to zero as more absorber is put in. Allowing for this background, the maximum range of the beta-particles is found to be 0.95 g/cm^2 of Al, corresponding, according to Feather's[7] formula, to a maximum energy of 2 mv. This value, in conjunction with the decay period, places the active body on Sargent's[8] upper curve.

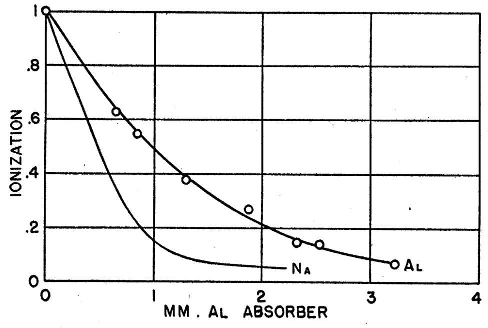

FIG. 3. Absorption of beta-particles from radio-aluminum in Al. Absorption of radio-sodium beta-particles shown for comparison.

The gamma-rays produce about 1/30 as much ionization as the beta-particles, which is the same ratio as was observed in the case of radio-sodium. This suggests, following Lawrence's[5] discussion of that case, that there is about one gamma quantum per beta-particle.

CHEMICAL IDENTIFICATION OF THE ACTIVE PRODUCT

The active product is presumably an element near Al, and most probably Mg, Al or Si. The possibility of Si was eliminated by dissolving an activated sample of thin Al foil in HF, evaporat-

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[5] E. O. Lawrence, Phys. Rev. **47**, 17 (1935).

[6] Curie, Joliot and Preiswerk (reference 10) have reported evidence for the occasional disintegration of Al^{28} with the emission of a neutron. We looked for this interesting effect with negative results. An activated Al target placed against a sheet of paraffin in front of a Wynn-Williams ionization chamber did not produce a detectable number of recoil proton counts, indicating that in this case less than 1 in 1000 of the disintegrating atoms emits neutrons. It should be pointed out that in the experiments of Curie, Joliot and Preiswerk the Al^{28} was produced by a different nuclear reaction and may have different properties.

[7] N. Feather, Phys. Rev. **35**, 1559 (1930).

[8] B. W. Sargent, Proc. Roy. Soc. **A139**, 659 (1933).

ing to dryness, and observing that the activity remained behind. Then a similar sample was dissolved in HCl, some $MgCl_2$ was added, and then an excess of KOH was added until the magnesium came down as a precipitate of $Mg(OH)_2$ while the aluminum remained in solution. The precipitate was found not to carry the activity. In a third experiment the sample was again dissolved in HCl, and precipitated with an excess of NH_4OH, which brings the aluminum down as $Al(OH)_3$. The activity was all found in the precipitate, showing definitely that the active product is an aluminum isotope. It is interesting to note that the same active substance has been produced by Curie and Joliot[9] on bombarding Mg with alpha-particles, by Curie, Joliot and Preiswerk[10] on bombarding P with neutrons, by Fermi and his co-workers[11] on bombarding Si with neutrons, and by Amaldi, D'Agostino and Segrè[12] on bombarding Al with neutrons.

EXCITATION FUNCTION OF THE RADIOACTIVITY

The number of radioactive atoms produced increases rapidly with the energy of the impacting deutons. The form of this variation was studied by using as a target a stack of thin Al foils (about 4 mm air equivalent stopping power each). Each foil receives precisely the same exposure to the deutons, with a difference in energy determined by the slowing down in the preceding foils. The measured activities of the foils are plotted against the corresponding deuton energies in Fig. 4. The circles are the experimental results, two different runs being shown on the plot.

This sort of experiment gives directly the differential excitation function. This can be compared to the theoretical curve of Gamow for the penetration of deutons through a nuclear potential barrier. In cases where the energy of the deutons is small compared to the height of the barrier, Gamow's formula states that the probability of penetration is proportional to

$$(1/v^2)e^{-4\pi Ze^2/hv} \qquad (1)^{13}$$

where v is the velocity of the deutons and Z the atomic number of the bombarded nucleus. This curve, for $Z=13$, and with the scale of ordinates adjusted to fit the higher experimental points, is shown in Fig. 4. It is seen that the fit with the experimental points is very good, except at the lower energies. This "tail" on the excitation curve may be due to a few faster deutons accompanying the main beam, or perhaps to a resonance penetration at low energies.

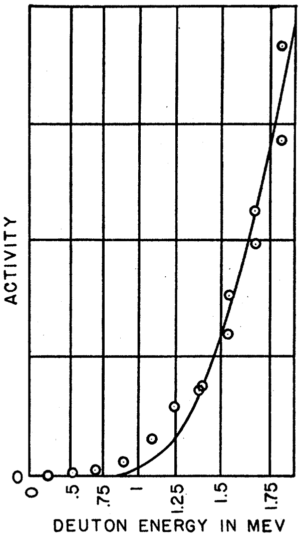

FIG. 4. Differential excitation curve of aluminum radioactivity.

NUCLEAR CROSS SECTION FOR ACTIVATION

The nuclear cross section for activation can be calculated when the form of the excitation function as well as the absolute intensity of activation of a thick target is known. The value obtained from the preceding data is 8×10^{-27} cm^2 at the maximum deuton energy of 1.9 mv.

RECOIL OF ACTIVATED NUCLEI

When a high speed deuton reacts with a nucleus, its momentum is imparted to the products of the reaction. The radioactive nucleus

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[9] I. Curie and F. Joliot, J. de phys. et rad. **5**, 153 (1934).

[10] I. Curie, F. Joliot and I. Preiswerk, Comptes rendus **198**, 2089 (1934).

[11] E. Fermi, E. Amaldi, O. D'Agostino, F. Rasetti and E. Segrè, Proc. Roy. Soc. **A146**, 483 (1934).

[12] E. Amaldi, O. D'Agostino and E. Segrè, La Ricerca Scientifica, November, 1934.

[13] Professor Oppenheimer informs us that there is an uncertainty as to the precise power of v in the factor multiplying the exponential. In Lawrence's paper on sodium $1/v$ is used.

receives most of this momentum plus a randomly directed momentum imparted by the other particle emitted in the reaction. Because of this randomness, the average momentum in the direction of motion of the deuton beam is just that imparted by the deuton. Dr. Kurie has kindly calculated that this gives the radioactive nuclei an average recoil range in the forward direction of about 0.6 mm of air for a deuton energy of 1.9 mv.

The existence of this recoil phenomenon has been noted previously,[14] and it has now been investigated more carefully in the case of aluminum. The experimental procedure was simply to replace the aluminum target by one of platinum, and to evacuate the space between the window and the target during the bombardment, so that the activated nuclei driven from the aluminum window by recoil would all be deposited on the platinum. It was found that the activity on the platinum was 1/20 as great as that on an aluminum target. From the form of the excitation curve it is calculated that this amount of activity is produced in a layer on the surface of the window of a thickness of 0.5 mm air equivalent. The depth of this layer is a fair measure of the average range of the recoil nuclei, and is in good agreement with the calculated value.

The Disintegration Protons

The fact that the radioactive product is an aluminum isotope, which decays with the emission of a negative electron, suggests that the nuclear reactions concerned are:

$$Al^{27}+D^2 \rightarrow Al^{28}+H^1 \qquad (2)$$

$$Al^{28} \rightarrow Si^{28}+e^-. \qquad (3)$$

The protons emitted in reaction (2) were investigated by the arrangement shown in Fig. 1, in which the target was in the vacuum. The protons emerged through a mica window and were counted by a Wynn-Williams counter—a shallow (4 mm) ionization chamber and linear amplifier in conjunction with a cathode-ray oscillograph and thyratron counter. Diaphragms in the path of the deuton beam and between the target and the counter insured that only particles from the target could reach the counter. Aluminum absorbers were placed in front of the counter, to obtain range curves. The deuton voltage in these experiments was 2.2 mv, and the current was usually kept small (about 0.1 μA).

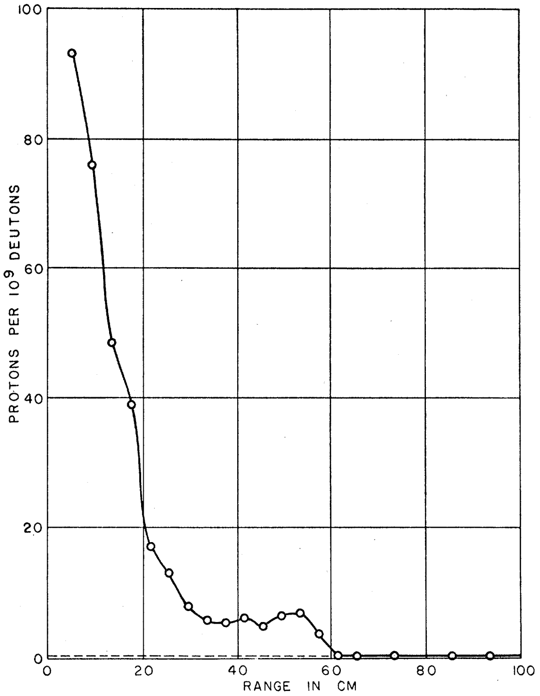

Fig. 5. Protons from a thick Al target bombarded with deutons. The counting rate (corrected to include the whole solid angle) is plotted against the air-equivalent thickness of absorber. The dotted line indicates the neutron background.

The first observations were made with a solid aluminum target, whose surface was carefully scraped to avoid contamination. The results are shown in Fig. 5 in which are plotted the numbers of protons emitted in all directions, calculated from the geometry of the apparatus. Each point represents about 500 proton counts. A diaphragm was put in front of the counter for the points below 20 cm, to obtain a reasonable counting rate. A small background of neutron counts is indicated by the dotted line.

It is known from other experiments (as pointed out by Lawrence[5] in his work on sodium) that a

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[14] M. S. Livingston and E. McMillan, Phys. Rev. **46**, 437 (1934). The nitrogen activity reported in this letter, which occurs as a contamination on any target bombarded with deutons in air, is not sufficiently large to interfere with the measurements on aluminum. In some of the experiments reported above, the space between the window and the target was evacuated during the bombardment in order to be sure that this contamination is of no importance.

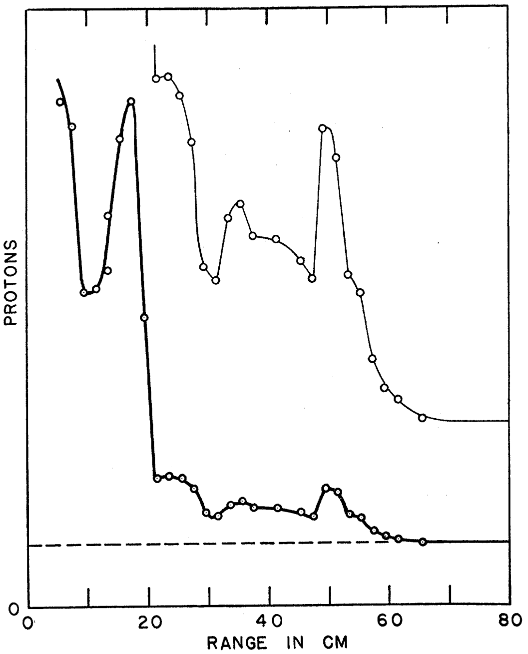

FIG. 6. Range distribution curve of protons from a thin Al target bombarded by deutons. The curve drawn with a finer line is the part beyond 20 cm with a larger scale of ordinates.

homogeneous group of protons, if present, is indicated by this counting arrangement as a sharp peak of half-width 2 cm. The question therefore arose as to whether these results were to be interpreted as showing a continuous distribution of proton energies, or a number of discrete groups that are too close together for resolution when using a thick target.

THIN-TARGET EXPERIMENTS

To settle the question of the nature of the proton distribution in energy, a target of thin Al foil was used (about 2 mm air equivalent thickness in the direction of the deuton beam). The results obtained in this way are shown in Fig. 6; the finer curve is simply the part beyond 20 cm plotted on a larger ordinate scale. As before, the readings below 20 cm were taken with a diaphragm in front of the counter, and each point represents about 500 proton counts. The number of proton counts is reduced by the use of the thin target, whereas that part of the neutron background not arising from the target is undiminished. Hence the neutron background appears relatively larger.

These results show that there are two very strong proton groups of 10 and 21 cm range, two weak but clearly resolved groups of 30 and 53 cm range, and still weaker groups between 30 and 48 cm as well as at least one more group with a maximum range of 62 cm. The ranges and relative intensities of the resolved groups, together with the reaction energies corresponding to the different ranges (obtained by allowing for the deuton energy and the recoil of the product nucleus), are summarized in Table I. The reaction energy of 5.3 mv presumably corresponds to the case in which the product nucleus is left in its lowest state, while in the others it is left in various excited states, with energies given by the differences in reaction energies. These excited nuclei should give rise to gamma-rays, evidence for which has been found by McMillan.[15]

TABLE I. *Proton ranges from radioactive aluminum.*

Range (cm)	Relative intensity	Reaction energy (mv)
10	7	0.6
21	6	1.8
30	1	2.9
53	0.8	4.6
62	—	5.3

YIELD OF PROTONS

The total number of disintegration protons can be found by integrating the curve of Fig. 5, using as unit abscissa interval the estimated half-width of a sharp proton peak, namely 2 cm. Such an integration, making a reasonable extrapolation to zero range, showed that there are 8 protons emitted in all directions per 10^7 deutons (of 2.2 mv energy) striking the target. The yield of radioactive atoms was found to be 3 per 10^7 deutons at 1.9 mv, a number which is in good agreement with the proton yield when the change in the reaction probability between 1.9 and 2.2 mv is allowed for. This gives further support to the view that the protons and the radioactivity arise from the same nuclear reaction.

NEUTRONS

The neutron background previously mentioned arises partly from the target and partly from other parts of the apparatus. It was investigated

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[15] E. McMillan, Phys. Rev. **46**, 868 (1934).

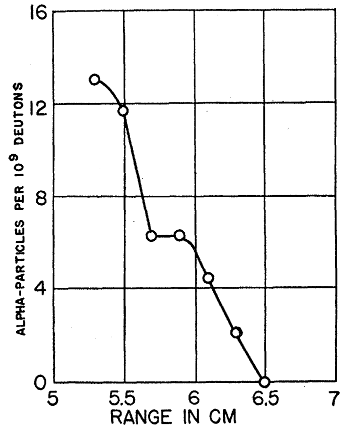

FIG. 7. Absorption curve of the alpha-particles from a thick Al target bombarded with deutons.

more carefully by placing a sheet of paraffin in front of the counter, and observing the number of counts with a thick Al target, and then with an inactive target in place. It was found that the counting rate was doubled by putting in the Al target. The total yield of neutrons from the Al calculated on the assumption that one per thousand traversing the counter was recorded, was 2 per 10^7 deutons. The reaction concerned is probably:

$$Al^{27}+D^2\rightarrow Si^{28}+n. \qquad (4)$$

ALPHA-PARTICLES

The counting circuit can be adjusted so as to record alpha-particles but not protons, and when this was done it was found that a few counts were still observed at short ranges. In this experiment the deuton current was made very small (0.007 μA) in order to avoid operation of the counter by the "piling up" of proton impulses, and the variation in range was obtained by moving the counter. The results, shown in Fig. 7, have been corrected for the variation in solid angle and are expressed in terms of the total number of alpha-particles emitted in all directions. The "step" in the curve was found in two separate runs, and is probably real.

Thus there appear to be two weak groups of alpha-particles, of ranges 5.7 and 6.5 cm, corresponding to reaction energies of 5.9 and 6.6 mv, respectively. The yield is about 6 per 10^9 deutons for each group, of the order of 1/100 as great as the proton yield. The reaction giving rise to the alpha-particles is:

$$Al^{27}+D^2\rightarrow Mg^{25}+He^4. \qquad (5)$$

EFFECTS WITH PROTON BOMBARDMENT

By admitting ordinary hydrogen to the apparatus instead of deuterium, a beam of hydrogen molecular ions (of course contaminated with a few deutons) was obtained. The proton energy in this case was 1.1 mv. By using a large current, a weak alpha-particle group was observed of the same range as found under deuton bombardment, accompanied by neutrons and fast protons, all in the same proportions as found with deutons. These effects were of a magnitude such as to be accounted for by a deuton contamination of one per two thousand protons. There is little doubt that this is the explanation, and that the protons themselves produce no appreciable number of disintegrations.[16]

ATOMIC MASSES

None of the masses concerned in any of the above reactions is known accurately from mass-spectrographic data, so that only differences can be computed. Assuming that the largest reaction energy in each case corresponds to the process which leaves the product nucleus in its normal state, and taking $H^1=1.0078$, $D^2=2.0136$, $He^4=4.0022$, we get:

From Eq. (2), $Al^{28}=Al^{27}+1.0001$;
From Eq. (5), $Al^{27}=Mg^{25}+1.9957$.

ACKNOWLEDGMENTS

The authors wish to thank Dr. M. Stanley Livingston for his collaboration in some of the radioactive work done while he was still in Berkeley. They also express their appreciation for the financial support given by the Research Corporation and the Chemical Foundation.

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[16] In the early experiments of M. S. Livingston and E. O. Lawrence (Phys. Rev. **43**, 369, 1933) the Geiger-counter observations of the radiations from Al bombarded by protons which were thought to be alpha-particles, were doubtless due to soft x-rays.