

# THE PHYSICAL REVIEW

*A Journal of Experimental and Theoretical Physics Established by E. L. Nichols in 1893*

VOL. 58, No. 10

NOVEMBER 15, 1940

SECOND SERIES

## The Disintegration of $N^{14}$ and $N^{15}$ by Deuterons

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(Received September 19, 1940)

A mixture of  $N^{14}$  and  $N^{15}$  gas has been bombarded with deuterons of 1 Mev and the disintegration particles observed at right angles to the direction of the deuteron beam. The ranges of the disintegration particles were measured by means of a shallow ionization chamber and pulse amplifier. The shortest measurable range was 2.1 cm (15°C, 760 mm Hg). The reaction  $N^{15}(d, \alpha)C^{13}$  gave rise to one group of alpha-particles at 5.25 cm range with a  $Q$  value of 7.54 Mev. Three groups of alpha-particles (11.97 cm, 6.54 cm and 3.47 cm range) were observed arising from the reaction  $N^{14}(d, \alpha)C^{12}$ . The  $Q$  values corresponding to these three groups are 13.39 Mev, 9.02 Mev and 5.77 Mev, respectively. From the reaction  $N^{14}(d, p)N^{15}$  two valid groups of protons were observed having ranges of 90.5 cm and 21.02 cm with  $Q$  values of

8.51 Mev and 3.15 Mev. The ranges given above are from a single measurement and the  $Q$  values are the average of the results of several measurements. A group at 66 cm previously reported due to this reaction was found to be due to boron contamination. No proton groups were found which could be attributed to the reaction  $N^{15}(d, p)N^{16}$ , although a radioactivity of half-life  $9.5 \pm 1.0$  sec. was found to result from deuteron bombardment of the mixture of  $N^{14}$  and  $N^{15}$ . This radioactivity was not found when  $N^{14}$  alone was bombarded. A method of range measurement is described which allows accurate corrections to be made for the effect of chamber depth and of the bias on the grid of the counting thyratron on the output of the pulse amplifier.

### INTRODUCTION

SINCE the careful work of Cockcroft and Lewis (1936)<sup>1</sup> very little work has been done on the measurement of the energy of the heavy particles emitted when nitrogen, carbon and oxygen are bombarded with high energy deuterons. Such heavy particle measurements are of particular interest in that they can give very accurate information about energy levels in the residual nuclei, although selection rules quite often prevent this method from giving information about all of the energy levels that one could on energy considerations expect to be involved. The present investigation is limited to a study of the  $(d, p)$  and  $(d, \alpha)$  reactions with  $N^{15}$  and

$N^{14}$  as the target nuclei. The results given in this report are based on more measurements with better corrections than those reported previously<sup>2,3</sup> and are believed to be more accurate.

### EXPERIMENTAL ARRANGEMENTS

The source of the high energy ions used in this experiment was the Cornell cyclotron. The beam of ions could be directed outside the cyclotron chamber to a position 14 to 30 cm from the chamber wall. As is shown in Fig. 1 the target cell is bolted to the exit port of the cyclotron. The deuterons enter the target cavity through an aluminum foil at the end of the foil tube.

<sup>1</sup> J. D. Cockcroft and W. B. Lewis, Proc. Roy. Soc. **154**, 261 (1936).

<sup>2</sup> M. G. Holloway and B. L. Moore, Phys. Rev. **56**, 705 (1939).

<sup>3</sup> M. G. Holloway, Phys. Rev. **57**, 347 (1940).

The foil is attached to the supporting gridwork with Glyptal. The target gas is admitted to the target cell through the tube at the left and the pressure of the gas is measured by means of a Bourdon type vacuum gauge which is not shown in the drawing. Such a gauge can be used in this experiment since the gas pressure in the target cell is usually less than 7 cm of Hg and errors in this measurement amount to only several hundredths of a millimeter range.

The disintegration particles to be observed pass out through the mica foil at the side of the target cell. The direction of these particles is generally at  $90^\circ$  to the direction of the deuteron beam, but no collimation is introduced at this point. After passing through the foil the disintegration particles traverse the range cell in which the pressure of dry air can be varied by means of an external system. This pressure is measured by a mercury manometer. The temperatures of the target cell, range cell, and ionization chamber are measured with three thermometers in copper tubes soldered to the respective parts. The particles enter the ionization chamber through a collimating head which supports an aluminum foil, which is one electrode of the parallel plane ionization chamber.

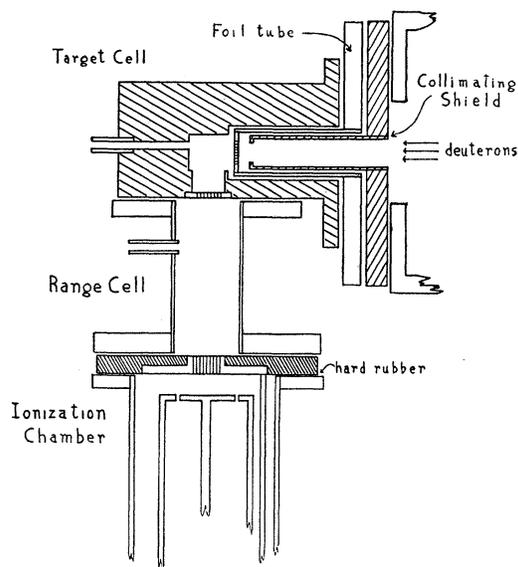


FIG. 1. Sketch of the assembly of target cell, range cell and ionization chamber used in measuring the ranges of the particles emitted in a direction  $90^\circ$  to the direction of the deuteron beam. The gaskets and screws which hold the components together are not shown.

The collecting electrode and its guard ring can be moved parallel to its axis on threads of 1-mm pitch turned on the guard ring cylinder. The physical depth of the ionization chamber is measured by moving the collecting electrode forward until it makes electrical contact with the collimating head and foil, then the guard ring cylinder is screwed back. The number of revolutions of this cylinder determines the physical depth of the ionization chamber. In some cases the ionization chamber was modified, but these modifications will be mentioned in connection with the particular experiment.

The collimating head on the ionization chamber allows a maximum angle of  $5.4^\circ$  with the direction of the axis of the range cell and ionization chamber. It is necessary to have such a collimator because for some of the short range measurements the physical separation of the two foils bounding the range cell was only 1.4 cm and the obliquity correction in this case was 0.5 percent and less in all other cases. All three gridworks were drilled from the same pattern and have a ratio of open to total area of 0.54.

The collecting electrode of the ionization chamber is connected directly to the grid of the first amplifier tube of the 6-stage linear pulse amplifier described previously.<sup>4</sup> The output of this amplifier is connected to the grid of a discriminating thyatron which operated a mechanical counter. This grid is biased so that the output voltage pulse must be above a certain value before the thyatron will flash. With no signal on the grid, the thyatron will flash at  $-15$  volts grid bias, so for any negative bias,  $V$ , greater than 15 volts, the pulse height required to flash the tube is  $V-15$  volts. It will be shown in a later section that the method of range measurement used in these experiments makes it necessary that the amplifier be linear in its response to voltage pulses. For this reason it was necessary to make frequent measurements of the response of the amplifier. This was done with the thyatron oscillator described in reference 4, p. 24.

It was necessary to use a current integrator because the deuteron beam was somewhat unsteady. This integrator is similar to the type

<sup>4</sup> M. G. Holloway and M. S. Livingston, *Phys. Rev.* **54**, 18 (1938).

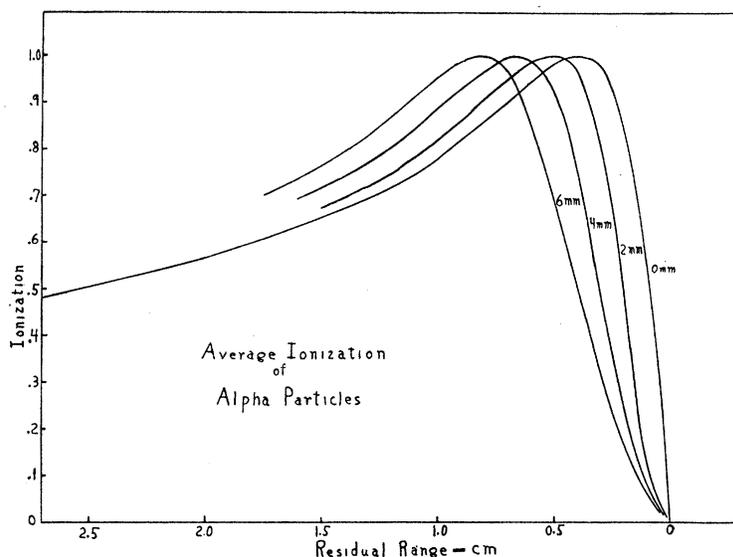


FIG. 2. The average ionization produced by alpha-particles in ionization chambers of different depths.

described by Herb, Kerst and McKibben<sup>5</sup> and has been found to be very satisfactory, although in the summer months precautions must be taken to prevent surface leakage across insulators. In Fig. 1 it can be seen that the grounded collimating shield prevents the beam from striking any part of the foil tube but the foil and supporting gridwork. Such a precaution is necessary, for if the beam shifts in position then a part of it will strike the solid edges of the supporting gridwork or the walls of the foil tube and no fraction of this current can reach the target gas although it will be measured by the integrator. However, with the beam restricted to the gridwork it is certain that 54 percent of the measured beam does get to the target gas.

For some of the longer range groups it would have been impractical because of intensity considerations to obtain sufficient absorber thickness by removing the ionization chamber a large distance from the target cell and consequently mica absorbers were used. A set of mica foils was made from *one* large piece of mica. The air equivalents of two thin pieces of this lot were measured with a collimated beam of polonium alphas; these foils were placed in the

alpha-particle beam so that the average energy of the alphas in the mica corresponded to a residual range of 3 cm of air. The areas and masses of the two pieces were measured and from these measurements the mass/cm<sup>2</sup>/cm air was found to be 1.44 mg/cm<sup>2</sup>/cm air and 1.41 mg/cm<sup>2</sup>/cm air; the average of these is 1.425 mg/cm<sup>2</sup>/cm air and this value was taken as the stopping power for that sample of mica for alphas of 3-cm residual range. From the mass/cm<sup>2</sup> of the thicker pieces their thickness in air centimeters could be obtained. However, some correction had to be made to the thicknesses so obtained because the factor 1.425 mg/cm<sup>2</sup>/cm air was obtained at a residual range of 3 cm and all of the thicker absorbers were used at much higher ranges. Livingston and Bethe<sup>6</sup> have given a table of the stopping power relative to air of aluminum for various velocities of alpha-particles and protons. The correction for mica was taken as half the correction for aluminum. For velocities above  $1.5 \times 10^9$  cm/sec. alpha-particles and protons have almost the same range. While these corrections are somewhat uncertain they are of second order in the range measurement and errors in the corrections are not too serious. The corrections for the one platinum foil used

<sup>5</sup> R. G. Herb, D. W. Kerst and J. L. McKibben, Phys. Rev. **51**, 691 (1937).

<sup>6</sup> M. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 272 (1937).

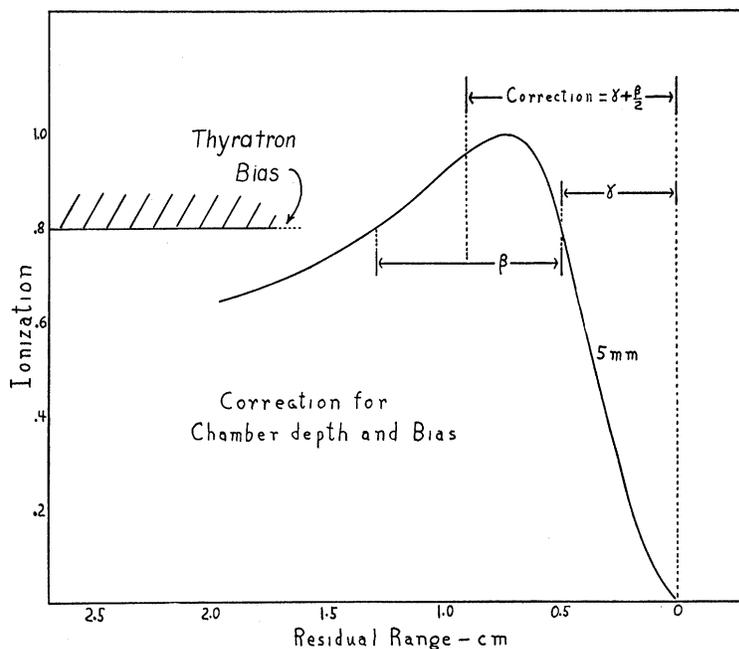


FIG. 3. Diagram showing the quantities involved in the correction to the range for ionization chamber depth and thyratron bias.

in these experiments were made with the curve for gold.

#### METHOD OF RANGE MEASUREMENT

The determination of the "range" in an absorber of a group of heavy particles is in most cases the problem of measuring the range of the group in such a manner that the average initial energy of the particles in the group can be determined; that is, a knowledge of the actual length of the path of a particle is useful only in that it is a means of obtaining the kinetic energy of the particle at some particular point. It is the purpose of this section to describe the method used in these experiments in obtaining the energy of the particle groups by means of range measurements. All ranges given in this paper are for dry air at 15°C and 760 mm of Hg.

Each alpha-particle or proton as it moves through a gas produces ion pairs along its path. The number of ion pairs per cm path length  $dI(x)/dx$  is not constant for any one particle, but is a decided function of the velocity of the particle. In air the dependence of  $dI(x)/dx$  upon velocity is such that  $dI(x)/dx$  has a maximum several millimeters from the end of its path.

This end of path is defined as that point along the path at which the particle has so little energy (<33 ev) that it cannot produce even one ion pair. In Fig. 2 is shown the ionization collected in parallel plate ionization chambers of different depths as a function of the distance of the face of the ionization chamber from the end of the path of an average alpha-particle. The differential specific ionization  $dI(x)/dx$  is given by the curve (0 mm) for zero chamber depth.<sup>7</sup> The chamber depth for each curve is indicated by the number to the right of the curve. These curves for chambers of finite depth are obtained from the differential specific ionization curve by taking the integral

$$\int_{x-\delta}^x \frac{dI(c)}{dc} dc,$$

where  $\delta$  is the chamber depth, and then normalizing the ordinate so that the maximum of the curve for each chamber depth is 1.0.<sup>8</sup>

<sup>7</sup> Reference 4, p. 29.

<sup>8</sup> From the work of I. Curie and F. Béhounek (J. de phys. et rad. 7, 125 (1926) and Ann. de physique 3, 299 (1925)) it can be deduced that the ion pairs corresponding to the maximum of the differential specific ionization curve

In Fig. 3 is shown a curve giving the ionization collected in a 5-mm chamber as a function of the distance of the face of the chamber from the end of the range of the alphas in the group having the mean range of the group. Since under rather wide operating conditions the voltage pulse on the grid of the first amplifier tube is proportional to the ionization produced by the individual alpha-particle in the chamber, the ordinate of the curve in Fig. 3 will also represent the voltage pulse on the discriminating thyratron as a function of the ion chamber position. This last is true only if the amplifier is linear in its response to voltage pulses. The drawing indicates a bias on the discriminating thyratron which would correspond to a pulse height 0.8 of the maximum average obtainable with the 5-mm chamber. With such a bias the chamber face could be at any position within  $\beta$  and the alpha-particle would still be counted by the discriminating thyratron. If the ion chamber face is anywhere outside of  $\beta$  the alpha would not be counted. If the range straggling is of the form  $\exp[-x^2/\alpha^2]$  it can be shown easily that the peak of the curve connecting number of counts with the position of the chamber face will be at the center of  $\beta$  or displaced by  $\gamma + \frac{1}{2}\beta$  from the mean range.  $\beta$  and  $\gamma$  are both functions of  $\delta$ , the chamber depth, and of  $P$ , the pulse height below which the discriminating thyratron will not flash.

For a very high bias corresponding to ionization pulses in the near neighborhood of 1.0,  $\beta(\delta, P)$  is very small and  $\gamma(\delta, P)$  is very nearly equal to the distance of the position of maximum ionization from the end of range. In this case the peak of the counting (high bias number-distance) curve will occur at the same position as that of the maximum of the ionization curve for that particular chamber depth.

It is very easy to realize experimentally these last conditions. The procedure is to take a number-distance curve and use a bias known from experience to be slightly higher than the mean pulse height at the maximum of the

ionization curve. The peak of this number-distance curve will be very near the position of the peak of the ionization curve; the accuracy of this being limited by the rather poor statistics coming from the few counts at high bias. The ion chamber is then placed at the position of maximum counting rate and a number bias curve taken (see Fig. 8, reference 4). At this position of the chamber the bias for half-maximum counting rate is then the bias corresponding to the mean of the ionization pulses arising from particles having their maximum ionization in the ion chamber. The bias of the discriminating thyratron is set at this bias which corresponds to ionization pulses of height 1.0 in Fig. 3. One must remember that the curve in this figure represents the ionization pulse such that half the particles in the group produce larger and half smaller pulses than that given by the ordinate of the curve. The number distance curve is then repeated with the bias corresponding to ionization pulse height 1.0. The true mean range of the group of particles is the sum of the distance between source and the position of the peak in the counting rate curve and of  $\gamma(\delta, 1.0)$  for the particular chamber depth used. If the bias is for a pulse height less than 1.0 then the correction is not  $\gamma(\delta, 1.0)$  but  $\gamma(\delta, P) + \frac{1}{2}\beta(\delta, P)$  appropriate for the particular bias. The reason for using a bias for pulse height as near 1.0 as possible is that this bias gives a larger ratio of maximum to half-width of the

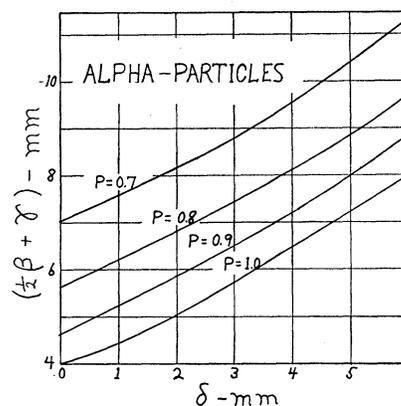


FIG. 4. The correction to the range of alpha-particles for thyratron bias and ionization chamber depth as a function of the chamber depth for different pulse heights (different thyratron biases).

is 6760 ion pairs/mm. With this value, the maxima of the curves for finite chamber depth are: 2 mm, 13,480; 4 mm, 26,400; 5 mm, 32,450; 6 mm, 38,400 ion pairs/mm. The numbers give the number of ion pairs produced in the chamber when the face of the chamber is set at the position of the maximum of the curve.

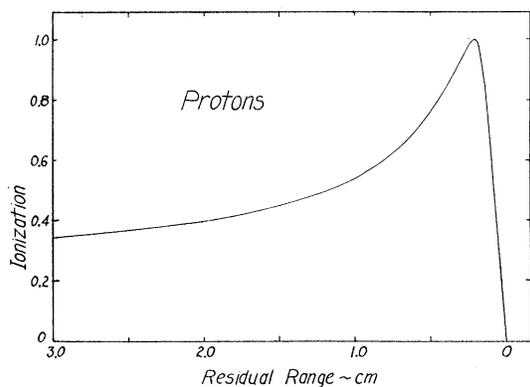


FIG. 5. The average differential specific ionization produced by a proton.

counting rate *vs.* position curve than any other bias.

From the set of curves in Fig. 2 the correction ( $\gamma + \frac{1}{2}\beta$ ) for different depths  $\delta$  and pulse height  $P$  can be obtained. Figure 4 gives this correction as a function of  $\delta$  for different  $P$ 's for alpha-particles. Again it should be mentioned that for most experiments  $P \cong 1.0$  and the range correction for bias and chamber depth is  $\frac{1}{2}\beta(\delta, 1.0) + \gamma(\delta, 1.0)$ ; for  $P < 1.0$ , the correction is  $\frac{1}{2}\beta(\delta, P) + \gamma(\delta, P)$ .

In order to check this method of range measurement the mean range of polonium alphas was measured with a 2-mm chamber and with a 4-mm chamber. The 2-mm chamber measurement gave 3.84<sub>5</sub> cm and the measurement with the 4-mm chamber gave 3.85<sub>5</sub> and 3.85<sub>0</sub> in two measurements. Since we have taken  $3.842 \pm 0.006$  cm as the standard range of the polonium alpha-group it would appear that this method of measurement gives the correct range to an accuracy limited by errors in measurement of the dimensions of the apparatus. The range and energy of the polonium alpha-group gives a point on the range-energy relation and thus, if the mean range of some new group of alpha-particles were measured by the method described above and found to be 3.84 cm one would be certain that the mean energy of the group is 5.29<sub>8</sub> Mev. Similar points between 2 Mev<sup>9</sup> and 10.5 Mev<sup>10</sup> based on measurements of the range and

<sup>9</sup> G. Mano, *Ann. de physique* **1**, 407 (1934); G. H. Briggs, *Proc. Roy. Soc.* **114**, 341 (1927).

<sup>10</sup> W. B. Lewis and B. V. Bowden, *Proc. Roy. Soc.* **145**, 235 (1934); S. Rosenblum and G. Dupouy, *J. de phys. et rad.* **4**, 262 (1933).

energy of alphas from naturally radioactive sources make the method reliable in this region. The theoretical extension of the range energy relation to energies higher than 10 Mev is straightforward and is quite accurate. The curve in the region below 2 Mev is obtained by integrating the differential specific ionization curve and fitting the integrated curve to the experimental curve at about 4 Mev; this may be in error because of a possible variation in energy loss per ion pair in the low energy region.

The measurement of the range of protons is a problem similar to that for alpha-particles. However, the differential specific ionization for an average proton of a group is not as well known as that of the alpha-particle. Parkinson, Herb, Bellamy and Hudson<sup>11</sup> in their work on the range of protons in air used a very shallow (0.04 cm) ionization chamber and obtained curves of ion current *vs.* energy (for a fixed air path) showing the familiar specific ionization curve for heavy particles. These curves of specific ionization *vs.* energy can be transformed into curves of specific ionization *vs.* range. In particular, curve VI, Fig. 5, reference 11, was so transformed. The transformed curve is a "Bragg" curve for a proton group having in this case a very small straggling. The effect of the finite chamber depth and range straggling is to put a small tail at the foot of the curve and to decrease the slope of the face of the specific ionization curve (this is discussed in reference 4). Initially it was assumed that the true differential specific ionization curve was the same as curve VI, Fig. 5, reference 11, except that the linear face of the specific ionization curves was extended directly to the axis, i.e., no tail. To this curve was then applied a straggling of 0.5 mm (more than would be expected from the Wisconsin high voltage generator) and a new curve computed. There was no appreciable change in the curve except that a tail was added to the foot and the curve displaced by about 1 mm. The effect of the finite chamber depth of 0.4 mm was also to add a tail and decrease the slope by about 15 percent. The differential specific ionization curve of the average proton of the group was then taken as curve VI, Fig. 5,

<sup>11</sup> D. B. Parkinson, R. G. Herb, J. C. Bellamy and C. M. Hudson, *Phys. Rev.* **52**, 75 (1937).

reference 11, with the linear face extrapolated to the axis and a 15 percent increase in slope of the face. This curve is shown in Fig. 5. In measuring ranges the same procedure as for alpha-particles is followed and the correction  $\frac{1}{2}\beta(\delta, P) + \gamma(\delta, P)$  for protons is given in Fig. 6. Because the curves for  $P=0.9, 0.8$  and  $0.7$  are so near the curve for  $P=1.0$ , only the correction for  $P=1.0$  is given. In as much as the proton produces only about a third the maximum ionization per millimeter as an alpha-particle it is very easy to distinguish between the two types of particles by the height of the voltage pulse which they produce in the output of the amplifier.<sup>12</sup>

The range-energy relation for protons was taken from the work of Parkinson, Herb, Bellamy and Hudson with some corrections to their data to get mean ranges and mean energies. Above 1.95 Mev the relation was obtained by transforming the alpha-particle range-energy relation by means of the relations

$$R_H(v) = (M_H/M_\alpha)(Z_\alpha/Z_H)^2 R_\alpha(v) - c$$

and

$$E_H(v) = (M_H/M_\alpha) E_\alpha(v).$$

The constant  $c$  takes care of the difference in the capture and loss effect between proton and alpha-particles, and is equal to  $0.3 \pm 0.06$  cm.

#### MEASUREMENT OF THE DEUTERON ENERGY

It was necessary to determine as accurately as possible the energy of the deuterons from the cyclotron, since the energy of the bombarding particle enters into the calculation of the disintegration energy of a nuclear reaction. Theoretically the energy can be calculated from the frequency of the oscillating "dee" voltage and the geometry of the cyclotron chamber. However, uncertainties in the determination of the radius

<sup>12</sup> For alpha-particles and protons of energies greater than 1 Mev and 0.1 Mev, respectively, the loss and gain of charge is negligible, and for alphas and protons of the same velocity the ratio of the differential specific ionization of the alpha-particle to that of the proton should be in the ratio of the square of their charges, namely 4/1. Comparison of the differential specific ionization curves of the alpha-particle and the proton in the region of 3-cm residual range gives an absolute ordinate for the proton curve such that the maximum corresponds to 2320 ion pairs/mm. If a very deep chamber is used it is necessary to extend the differential specific ionization curve to greater residual ranges. This can be done by using Eq. (749), reference 6.

of curvature of the last half-circular path make this method unsatisfactory for accurate work. A more direct method is to measure the range of the particles, but because of the large beam intensity a pulse amplifier could not be used to count the individual particles and as a result the correction for chamber depth could not be made accurately. This inaccuracy in the determination of the range of the very short range deuterons was too large to allow the use of this method.

A third method, which was the one adopted in the present work, is to use some nuclear reaction. The one chosen here was the reaction  $H^2(d, p)H^3$ . If the  $Q$  value of this reaction and the energy of the emitted proton (say at  $90^\circ$ ) is known the energy of the bombarding particle can be computed. However, by measuring the energy of the protons emitted in the forward direction and at  $90^\circ$  to the deuteron beam it is possible to determine both the  $Q$  value and the energy of the incident deuterons. Once the  $Q$  value is known it is necessary to measure the proton range only in the  $90^\circ$  direction in order to obtain the deuteron energy.

The experimental arrangement for measuring the protons in the  $90^\circ$  direction was the same as that previously described with a 19.2-cm range cell. For the measurement at  $0^\circ$  a 33-cm range cell and a target cell with a foil on the forward end was used. The depth of the target cell was 1.1 cm. Since the energy of the deuterons incident on the target gas was about 1 Mev and

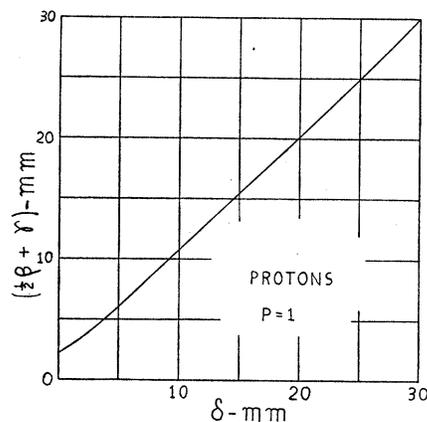


FIG. 6. The correction to the range of protons for ionization chamber depth for a thyratron bias corresponding to  $P=1.0$ .

the air equivalent of the gas was about 0.1 cm there was left about 1.4 cm of the deuteron range to be stopped in the exit foil. At this bombarding energy there is a group of protons from aluminum of about 35-cm range which might have interfered with the measurement of the group from the deuterium. In order to eliminate this the last foil was made up of a platinum foil of 1.1 cm air equivalent and an aluminum foil of 0.46 cm air equivalent. This last foil was not entirely made of platinum because the correction of the stopping power of platinum for different velocities is not very well known, and it was desirable to keep this correction as small as possible. The excitation function for the aluminum reaction becomes very small for 0.4-Mev deuterons and no protons from the small thickness of aluminum were observed in the region of 33-cm range.

Equations (1) and (2) relate the  $Q$  value and the deuteron energy  $E_1$ , with the energies of the protons emitted in the  $\underline{1}$  forward and  $90^\circ$  directions.<sup>13</sup>

$$E_1 = \frac{(M_2 + M_3)^2 (E_2^0 - E_2^{90})^2}{4M_1M_2 E_2^0}, \quad (1)$$

$$Q = \frac{M_2 + M_3}{M_3} \left[ E_2^{90} - \frac{(M_3 - M_1)(M_3 + M_2)}{4M_1M_2} \times \frac{(E_2^0 - E_2^{90})^2}{E_2^0} \right]. \quad (2)$$

The following notation is used: The subscript 0, 1, 2 and 3 refer to the initial nucleus, incident particle, emitted particle and residual nucleus, respectively. The superscript 0 will refer to the measurement in the  $0^\circ$  direction and 90 to the

<sup>13</sup> From conservation of mass and energy  $Q = E_2 + E_3 - E_1$  and from the conservation of momentum

$$M_3E_3 = M_1E_1 + M_2E_2 - 2(M_1M_2E_1E_2)^{\frac{1}{2}} \cos \theta,$$

where  $\theta$  is the angle between the direction of the incident and emitted particles. Measurements of the energy of the protons emitted in the forward direction and in the  $90^\circ$  direction allow the use of two independent equations involving the unknown quantities  $Q$  and  $E_1$ .

$$(a) \quad Q = \frac{M_2 + M_3}{M_3} E_2^{90} - \frac{M_3 - M_1}{M_3} E_1 \text{ at } 90^\circ,$$

$$(b) \quad Q = E_2^0 + \frac{[(M_1E_1)^{\frac{1}{2}} - (M_2E_2^0)^{\frac{1}{2}}]^2}{M_3} - E_1 \text{ at } 0^\circ.$$

From these two equations can be obtained the Eqs. (1) and (2) above.

$90^\circ$  direction. The cyclotron was not shut down during the change of target cells so that conditions in the cyclotron would be the same for the two measurements. The mean range of the protons in the forward direction was 30.63 cm corresponding to an energy of 4.71<sub>5</sub> Mev; in the  $90^\circ$  direction the range and energy was 15.94 cm and 3.22 Mev, respectively. Using these values of  $E_2^0$  and  $E_2^{90}$  in Eqs. (1) and (2) one is led to the value 0.95 Mev for the energy of the deuterons in the center of the gas in the target cell and 3.98 Mev for the  $Q$  value for the reaction  $H^2(d, p)H^3$ .<sup>14</sup> Since the air equivalent of the aluminum foil through which the deuterons passed into the target cell was 0.80 cm the energy of the deuterons incident on the foil was 1.27 Mev. From this energy value and the frequency of the cyclotron oscillator the radius of the last circle in the cyclotron can be computed and used for future determinations of the deuteron energy. However, this must be checked frequently because adjustments of the cyclotron can very easily change the geometry enough to require a new calibration of the beam energy.

In order to see how errors in  $E_2^0$  and  $E_2^{90}$  affect  $E_1$  and  $Q$  we take the differentials of Eqs. (1) and (2).

$$\delta E_1 = \frac{(M_2 + M_3)^2}{4M_1M_2} \left[ \frac{2(E_2^0 - E_2^{90})}{E_2^0} \delta(E_2^0 - E_2^{90}) - \left( \frac{E_2^0 - E_2^{90}}{E_2^0} \right)^2 \delta E_2^0 \right], \quad (3)$$

$$\delta Q = \frac{M_2 + M_3}{M_3} \delta E_2^{90} - \frac{(M_3 - M_1)}{M_3} \delta E_1. \quad (4)$$

The first item in (3) involves inaccuracies in the *difference* ( $E_2^0 - E_2^{90}$ ) and the second term inaccuracies in  $E_2^0$  alone, and since the errors in the two terms are approximately independent of each other the two terms must be added as the square root of the sum of their squares.

In the case of the measurement of the beam energy by the method of observing the emitted protons in two directions from the disintegration

<sup>14</sup> M. L. E. Oliphant, A. R. Kempton and Lord Rutherford, Proc. Roy. Soc. **149**, 406 (1935), give  $3.98 \pm 0.02$  Mev and F. E. Myers, R. D. Huntoon, C. G. Shull and C. M. Crenshaw, Phys. Rev. **56**, 1104 (1939), give  $3.93 \pm 0.10$  Mev.

of deuterium, the peak of the counting curve could be determined to about 2 mm of Hg pressure and the temperature to about 0.5°C. The estimated error in the measurement of the length of the 19.2 cm long range cell was about 0.01 cm. These errors give together a total error in the air equivalent of the range cell of 0.06 cm of air. There are two foils whose air equivalents are known to about 0.01 cm. The combined error in the chamber depth measurement and the chamber depth corrections to the range is about 0.02 cm. These constitute the experimental errors in the range of the 15.94-cm protons (90°) and total about 0.065 cm error for the 19.2-cm range cell. The longer range cell of length 33.0 cm has a somewhat larger error amounting in all to 0.08 cm in the 30.63-cm group observed at 0°. The difference ( $E_2^0 - E_2^{90}$ ) does not involve the inaccuracy in one foil or the error in the chamber depth and its corrections, and consequently the error in this difference is 0.094 cm rather than 0.103 cm as would be expected from adding the errors in the two ranges. The above errors are all the result of inaccuracies in measuring physical quantities in the course of the experiment.

The proton range-energy relation of Parkinson, Herb, Bellamy and Hudson includes the region up to about 2 Mev and is accurate to about 1 percent in range and energy. As was mentioned previously the range-energy relation for protons above 2.0 Mev is obtained by transforming the relation for alpha-particles. This includes the additive constant  $c$  which comes out to be 0.3 cm. By taking the probable errors in the alpha-particle curve and that of Parkinson, Herb, Bellamy and Hudson, the error in  $c$  is found to be 0.06 cm. Other errors involved in the absolute range scale for the protons are small compared to this error of 0.06 cm in  $c$ . However, for *differences* on the range energy relation for protons the accuracy is much better being about 0.02 cm.

The combination of the experimental errors and the errors in the range-energy relation gives the following errors for the ranges of the protons from the reaction  $H^2(d, p)H^3$ : In the forward direction, 0.10 cm and in the 90° direction, 0.09 cm. These correspond to inaccuracies in the energy of  $E_2^0$  of 11 kev and in  $E_2^{90}$  of 10.8 kev, the slope of the range energy relation in these

regions being 115 kev/cm. The difference ( $E_2^0 - E_2^{90}$ ) does not involve the absolute error in the range-energy relation, but rather a smaller error of about 0.02 cm. The error in the energy difference is then about 5 kev.

Experimentally it was not possible to determine the direction of the deuteron beam to better than 2°, and as a consequence the energy values 4.71<sub>5</sub> Mev and 3.22 Mev, while being accurately the energy of the protons observed, may not be  $E_2^0$  and  $E_2^{90}$  since these two quantities are defined as the proton energies in exactly the 0° and 90° directions. The inaccuracy in  $\theta$  introduces an error in  $Q$  of 72 kev and in  $E_1$  of 52 kev. These errors are large compared to the errors arising from the range measurements, and are connected only with the determination of the beam energy and the  $Q$  value, and not with the method used in determining the energy of the emitted particle. The errors combine to give an error in  $Q$  of 74 kev and in  $E_1$  of 54 kev.

## RESULTS

The target gases used in these experiments contained a high percentage of the heavy isotope N<sup>15</sup>. Some of the earlier work was done with a sample containing 20 percent of the N<sup>15</sup> and the later work with a 70-percent sample. The appreciable amount of the N<sup>14</sup> in all of the samples made necessary the study of the normal nitrogen under deuteron bombardment so that the contribution of the rarer isotope could be determined. This work on normal nitrogen was essentially a repetition of the work of Cockcroft and Lewis, but since the measurements on N<sup>14</sup> had to be made it was thought worth while to make them carefully.

In Fig. 7 are shown two groups of alpha-particles resulting from the bombardment of tank nitrogen which contains only about 0.4 percent of the N<sup>15</sup>. The reaction involved is  $N^{14}(d, \alpha)C^{12}$ . In view of some work to be mentioned later the small hump occurring at about 5 cm range is probably due to the presence of the small amount of N<sup>15</sup> in the tank nitrogen. The long range group at about 11.67 cm was measured in two ways.<sup>15</sup> The first measurement

<sup>15</sup> The range scale for the curves of counting rate vs. distance from the target cell is not absolute, but may differ by as much as 1 mm from the absolute value. The

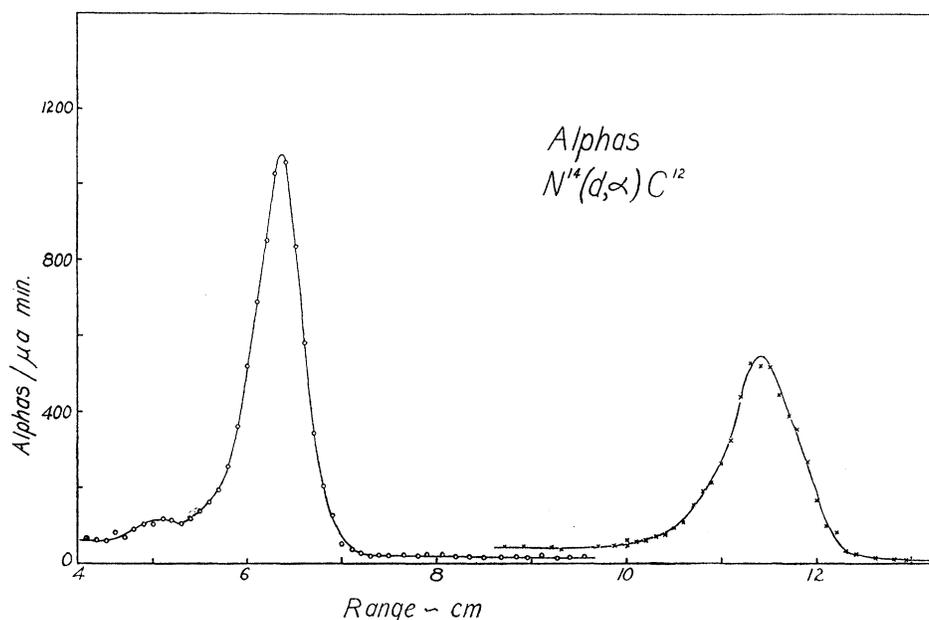


FIG. 7. Two groups of alpha-particles from the reaction  $N^{14}(d, \alpha)C^{12}$ . The target was tank nitrogen.

was with a foil holder between the range cell and the ionization chamber and with a copper screen as the face of the ionization chamber. The end of the range cell was covered in this case with a mica foil of about 0.88 cm air equivalent. A thick mica foil of 4.50 cm air equivalent from the calibrated set mentioned before was in the foil holder. With corrections for the stopping power of the mica and for slight obliquity the range of the group was 11.67 cm air, the bombarding energy being 1.01 Mev. The second measurement was with the 19.2-cm range cell used previously in measuring the range of the protons from the reaction  $H^2(d, p)H^3$ . There was no thick absorbing foil used in this measurement and the ionization chamber face was the Al foil over the collimating gridwork. The range found by this measurement was 11.97 cm at

accurate ranges are obtained from the counting rate vs. pressure curves. It would be much too laborious to transform each experimental point to an absolute scale, for the corrections required to get an accurate range scale are the following: (a) The contribution of the target cell must be corrected for pressure and temperature, (b) the range cell must have a similar correction, (c) the foils on the ends of the range cell as well as any thick absorbers must be corrected for the change in stopping power with velocity, (d) the chamber depth must be computed and the appropriate correction made, and (e) the obliquity factor computed and applied to the range. The ordinates of these curves do not represent absolute yields.

1.02-Mev deuteron energy. The  $Q$  values for these two measurements are 13.30 Mev and 13.48 Mev, respectively; the average of these is 13.39 Mev as compared with the mass  $Q$  value of 13.36 taken from the mass tables of Barkas.<sup>16</sup>

The group at 6.54 cm range (deuteron energy 1.02 Mev) is also due to the reaction  $N^{14}(d, \alpha)C^{12}$  in which the  $C^{12}$  is left in an excited state. The average  $Q$  value as taken from 5 separate measurements was 9.02 Mev. The two different ionization chamber heads and range cells used for 11.7-cm group were used in getting the 5 measurements of this group, although no thick mica absorber was involved.

The 6.54-cm group is shown again in Fig. 8 along with the short range group of alpha-particles at 3.47 cm, the deuteron energy being 1.02 Mev. Since these curves were taken with only tank nitrogen in the target cell it is probable that this group is from the  $N^{14}$ . Checks with the target cell evacuated and with  $CO_2$  in the cell indicate that it is not from a casual contaminant. The fact that its intensity was not enhanced by the presence of  $N^{15}$  in large quantities makes it likely that it does not arise from  $N^{15}$ . This group at 3.47 cm appeared with the

<sup>16</sup> W. H. Barkas, Phys. Rev. 55, 691 (1939).

$N^{14}$ – $N^{15}$  mixture, and with tank nitrogen in 5 distinct observations which makes it very probable that it is due to the  $N^{14}$  reaction. The average  $Q$  value from two range measurements is 5.77 Mev. Cockcroft and Lewis did not find this group of alphas, but an inspection of their published curves shows no experimental points in the region of this small peak.

In Fig. 9 are shown the alpha-particle groups obtained in the region 2- to 8-cm range when the target cell contained the gas mixture of 70 percent  $N^{15}$  and 30 percent  $N^{14}$ . In addition to the two groups at 6.54 cm and 3.47 cm there is a strong group at 5.25 cm; these ranges are for a bombarding energy of 1.02 Mev. The average of three range measurements on this group with slightly different deuteron energies gives a  $Q$  value of 7.54 Mev. This must correspond to the transition to the ground state of  $C^{13}$  in the reaction  $N^{15}(d, \alpha)C^{13}$ , for no alpha-groups of longer range were found which could be attributed to the presence of  $N^{15}$  in the target. The  $Q$  value as computed from Barkas' mass values is 7.51 Mev.

The very intense distribution of alphas below about 3-cm range was observed by Cockcroft and Lewis. The shortest range which could be studied by the present apparatus was about 2.1 cm, consequently any definite information concerning the short range distribution must be left for future measurements.

In the first report of this work<sup>2</sup> three groups of protons were attributed to the reaction  $N^{14}(d, p)N^{15}$ . The ranges of these groups were

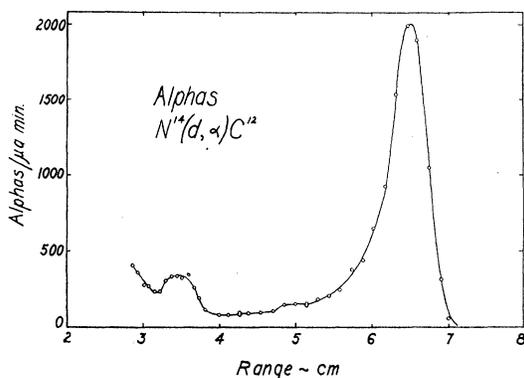


FIG. 8. Alpha-particle groups from the reaction  $N^{14}(d, \alpha)C^{12}$ . The group at about 6.5 cm is the same group shown in Fig. 7. The target was tank nitrogen.

given as 90.8 cm, 66.1 cm and 21.0 cm. The statistics in the early work were very poor in the case of the 90.8-cm group because of its small intensity compared to the neutron recoil background. Recently it was thought advisable to repeat the measurement of the 90-cm group with an ionization chamber of 1-cm physical depth filled with krypton at two atmospheres. The ionization produced in this chamber by a neutron recoil was small compared to the ionization from a proton which went completely through the chamber. With the high thyratron bias method very few of the neutron recoils were counted. The 90-cm group is shown in Fig. 10. The small dotted curve is the one taken with the 5-mm ionization chamber filled with air. The thick mica absorbers used in this measurement amounted to 71.6 cm of air, including the correction for the change in stopping power with velocity. Rather than make corrections for the large chamber depth which would of necessity

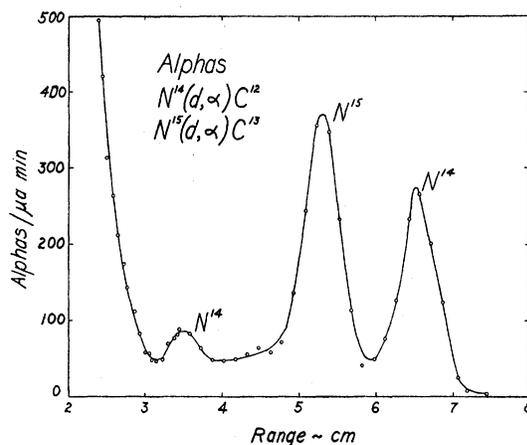


FIG. 9. Alpha-particle groups from a target of 70 percent  $N^{15}$ . The group at about 5.2-cm range is from the reaction  $N^{15}(d, \alpha)C^{13}$ . The other two groups were seen previously to be from the reaction  $N^{14}(d, \alpha)C^{12}$ .

require a knowledge of the stopping power of krypton the ionization chamber was calibrated by measuring the ranges of the protons for the reactions  $H^2(d, p)H^3$  and  $C^{12}(d, p)C^{13}$ . These two groups have a range of about 16 cm and have been carefully measured in this laboratory with the thin ionization chamber. The range of the group from the reaction  $N^{14}(d, p)N^{15}$  as measured with the thick ionization chamber is 90.5 cm. The older measurements with the thin ionization

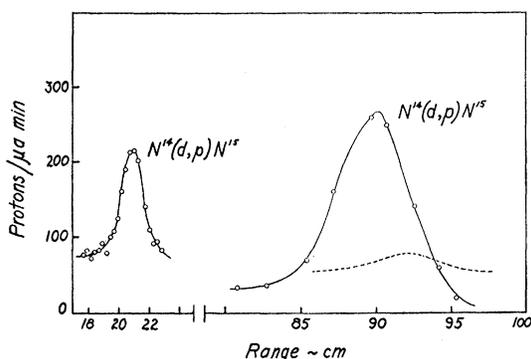


FIG. 10. The two groups of protons from the reaction  $N^{14}(d,p)N^{15}$ .

chamber gave 92.5 cm when corrected for change in stopping power of mica with velocity although with a different deuteron energy. Because of the much better statistics it is thought that the 90.5-cm range is the better value, and the  $Q$  value obtained with this range is 8.51 Mev as compared with the mass  $Q$  value of 8.57 Mev.

In the early work with the thin ionization chamber the proton group at 66 cm was more intense than the 90-cm group. However, with the thick krypton chamber the 66-cm group could not be found at all! Various common possible contaminants were tried but none gave the 66-cm group. Finally because the range of the group was about that to be expected from boron a target of borax was tried, and the 66-cm group appeared with a very large intensity. The problem was then to find the source of the boron in the earlier experiments and why the group did not show up when the target had been evacuated or filled with carbon dioxide but was present when tank nitrogen or the  $N^{14}-N^{15}$  mixture was used. It was remembered that a Bourdon type vacuum gauge had been used on the target cell for testing the cell for leaks and that this gauge had been discarded because of a leak in its flattened tube. Further investigation revealed that this gauge had been used several years previously in the filling of boron trifluoride ionization chambers, and had been laid aside because of the leak. This explained the source of the boron in the early experiments, for presumably the cavity into which the gauge had been screwed had become saturated with the gas. The time required for taking the points for one

of the long range proton groups was of the order of several hours and the  $N^{14}-N^{15}$  mixture or the tank nitrogen might remain in the target cell for as much as a day. This would allow ample time for some of the boron trifluoride to seep into the bombarding cavity. However, when the tests were made with the cell evacuated or with carbon dioxide very little time was required, since the method of checking the groups was to set the ionization chamber at the position of the peak of the curve and measure the counting rate. If the counting rate was the same as the background it was assumed that the group did not come from the cell walls or the carbon dioxide. In the time required for the check it is probable that very little of the boron trifluoride got into the target cell.

The strong group of protons at 21.02 cm (deuteron energy of 1.01 Mev) is from the reaction  $N^{14}(d,p)N^{15}$ . The  $Q$  value is 3.15 Mev which indicates an excitation level in  $N^{15}$  of 5.42 Mev.

A group of protons might be expected to result from the reaction  $N^{15}(d,p)N^{16}$ . The residual nucleus  $N^{16}$  is known to be radioactive with a half-life of 8 sec. Naidu and Siday<sup>17</sup> measured about 240 tracks in the beta-ray spectrum of  $N^{16}$  and obtained a visually extrapolated end point at little over 6 Mev. Fowler, Delsasso and Lauritsen<sup>18</sup> found a visual end point of about 6 Mev and a K-U limit of 6.5 Mev. If 6.5 Mev is taken as the energy of the beta-particles given off from  $N^{16}$  in the disintegration  $N^{16} \rightarrow O^{16} + e^-$  the mass of  $N^{16}$  has the lower limit of 16.007 MU and may be heavier by the energy of any gamma-rays involved in the disintegration. In the present experiment a thin-walled Geiger counter tube was set up very near to the foil window of the target cell. A chronograph was used to record every sixteenth count of the Geiger counter. With the target cell filled with the  $N^{14}-N^{15}$  mixture two half-life measurements were taken of the activity produced in the gas. The first measurement showed an 8.1-sec. half-life superimposed on a 5.6-min. half-life. The second measurement gave a 10-sec. half-life on

<sup>17</sup> R. Naidu and R. E. Siday, Proc. Phys. Soc. London **48**, 332 (1936).

<sup>18</sup> W. A. Fowler, L. A. Delsasso and C. C. Lauritsen, Phys. Rev. **49**, 561 (1936).

a 7-min. half-life. It was determined that the longer periods came from the cyclotron chamber and not from the target cell and presumably this longer half-life radioactivity came from the  $N^{13}$  formed in the cyclotron chamber from carbon contamination. The somewhat small half-life value for the  $N^{13}$  is due probably to the diffusion of the radioactive nitrogen from the walls into the pumping system. With the target cell filled with tank nitrogen there was no short life component found at all, but rather one of 6.7 min. half-life, which again was found to come from the chamber. Since  $N^{16}$  has a half-life of 8 sec.<sup>19</sup> the short-lived activity detected as coming from the bombardment of the  $N^{15}$  with deuterons is probably from  $N^{16}$  formed in the reaction  $N^{15}(d, p)N^{16}$ . If this is assumed to be the case then the fact that it is possible to produce the above reaction with 1-Mev deuterons sets an upper limit of 16.0126 MU for the mass of  $N^{16}$ . This value is for zero energy of the proton which is given off in the deuteron reaction. If the value 16.007 is used for the mass of  $N^{16}$ , the  $Q$  value for the reaction  $N^{15}(d, p)N^{16}$  is 4.19 Mev and with 1-Mev deuterons the energy of the emitted proton should be 4.75 Mev or a range of 31 cm of air. The entire region from 2-cm to 100-cm range was searched for protons with the  $N^{14}$ - $N^{15}$  gas mixture in the target cell, and there were found 3 groups of protons at 21 cm, 66 cm and 90 cm; these groups were also found with the tank nitrogen and their source is known. No new proton group was found. Below 12-cm range it was very difficult to detect protons because of the large numbers of alpha-particles in that region. Figure 11 shows a typical curve taken with a low bias for detecting protons. Since the weak group of alphas at 3.5 cm does not show on this curve, it is evident that a proton group would have to be rather strong to be detected over the strong background of alpha-particles. With only the results of the present experiment the matter of the protons from the reaction  $N^{15}(d, p)N^{16}$  must be left in a rather inconclusive state.

In Table I the results of all these measurements are summarized.  $E_1$  is the energy of the deuterons in the center of the gas target;  $E_2$  is the energy

of the emitted particle whose range in air (at 15°C, 760 mm Hg) is given in the third column.  $Q_{exp}$  is the  $Q$  value as computed from the present measurements and in the next two columns are given the  $Q$  values from Cockcroft and Lewis and from the mass tables of Barkas. The absolute values of the cross sections are probably accurate to a factor of 5, but the relative values are good to 50 percent.

The agreement between the  $Q$  values found in the present work and those of Cockcroft and Lewis is good except in the case of the alpha-group corresponding to the transition to the ground state of  $C^{12}$ ; the difference is 0.17 Mev, which is within the experimental errors given. The agreement is excellent in every case with the corrected values deduced from Cockcroft and Lewis' experiments by Livingston and Bethe.<sup>20</sup> The experimental  $Q$  values reported here and the  $Q$  values obtained from mass values agree to at least 60 kev.

Consideration of the energy levels shown in the table would lead one to expect that the bombardment of nitrogen with deuterons would produce gamma-rays having energies 4.37, 7.62, 5.42 and 3.25 Mev; the last gamma would come from a transition between the 7.62- and 4.37-Mev levels in  $C^{12}$ . Gaerttner and Pardue<sup>21</sup> have observed five gamma-ray lines resulting from the bombardment of nitrogen with deuterons; the energies of these lines were given as 2, 4,  $5.3 \pm 0.4$ ,  $7.2 \pm 0.4$  and 11 Mev. The 4- and 7.2-Mev lines could correspond to the 4.37- and

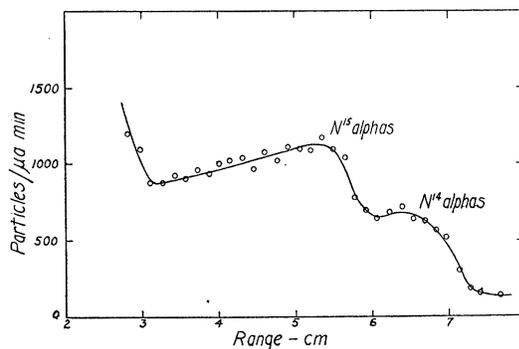


FIG. 11. Curve taken in the search for protons of range less than 8 cm.

<sup>19</sup> J. J. Livingood and G. T. Seaborg, Rev. Mod. Phys. 12, 30 (1940).

<sup>20</sup> Reference 6, pp. 279, 321 and 327.

<sup>21</sup> E. R. Gaerttner and Louis A. Pardue, Phys. Rev. 57, 386 (1940).

TABLE I. Data on the bombardment of nitrogen by deuterons.

REACTION	$E_1$ MEV	RANGE AIR CM	$E_2$ MEV	$Q_{\text{EXP}}$ MEV	$Q_{\text{C. \& L.}}$ MEV	$Q_{\text{MASS}}$ MEV	ENERGY LEVEL MEV	CROSS SECTION CM <sup>2</sup>
$\text{N}^{14}(d, \alpha)\text{C}^{12}$	1.01	$11.67 \pm 0.11$ $11.97 \pm 0.07$	$10.60 \pm 0.07$ $10.76 \pm 0.06$	$13.39 \pm 0.08$	$13.22 \pm 0.1$	$13.36 \pm 0.16$	$\text{C}^{12}$ g.s.	$1 \times 10^{-27}$
	1.01	6.39	7.31					
	1.03	6.57	7.45					
	1.02	$6.54 \pm 0.06$	$7.43 \pm 0.05$	$9.02 \pm 0.07$	$8.93 \pm 0.03$		$\text{C}^{12}$ 4.37	$3 \times 10^{-27}$
	1.03	6.49	7.40					
	1.02	6.47	7.39					
$\text{N}^{15}(d, \alpha)\text{C}^{13}$	1.03	$3.50 \pm 0.07$	$4.98 \pm 0.06$	$5.77 \pm 0.07$			$\text{C}^{12}$ 7.62	$3 \times 10^{-28}$
	1.02	$3.47 \pm 0.07$	$4.96 \pm 0.06$					
$\text{N}^{15}(d, \alpha)\text{C}^{13}$	1.01	$5.10 \pm 0.07$	$6.37 \pm 0.06$			$7.51 \pm 0.28$	$\text{C}^{13}$ g.s.	$1 \times 10^{-27}$
	1.02	$5.25 \pm 0.07$	$6.48 \pm 0.06$	$7.54 \pm 0.07$				
$\text{N}^{14}(d, p)\text{N}^{15}$	0.95 (1.01)	$90.5 \pm 1.6$ (92.5)	$8.75 \pm 0.1$ (8.86)	$8.51 \pm 0.1$ (8.57)	$8.53 \pm 0.1$	$8.57 \pm 0.23$	$\text{N}^{15}$ g.s.	$4.5 \times 10^{-28}$
	1.01	$21.02 \pm 0.11$	$3.78 \pm 0.02$	$3.15 \pm 0.05$	$3.25 \pm 0.07$		$\text{N}^{15}$ 5.42	$3 \times 10^{-27}$

7.62-Mev levels in  $\text{C}^{12}$  and the 5.3-Mev line with the 5.42 level in  $\text{N}^{15}$ . The relative intensities of the  $\alpha$ -particle transitions to the 4.37-Mev level in  $\text{C}^{12}$  and the 5.42-Mev level in  $\text{N}^{15}$  agree approximately with the relative intensities of the 4- and 5.3-Mev gamma-ray lines. However, it should be pointed out that the ratio of the numbers of the particle transitions to the 4.37- and 7.2-Mev levels in  $\text{C}^{12}$  is 10 whereas the ratio of the numbers of the 4- and 7.2-Mev gamma-ray quanta as given by Gaertner and Pardue is 0.45. In order to explain this one might assume the existence of another strong group of alphas corresponding to a still higher excited state of  $\text{C}^{12}$ . Such a group would of course fall into the very short range region and could not be detected in the present measurements. The corresponding excited state of  $\text{C}^{12}$  would be unstable against alpha-emission, but it is still easily conceivable that such a state could not actually emit alpha-particles because of selection rules. If one assumes that such a state exists at about 9 Mev it is easy to account for the intensity of the 7.2-Mev line as well as the existence of the 2-Mev

line which could originate in the transition from the 9 Mev to the 7.62-Mev level. This picture is given some support by the fact that the intensities of the 2-Mev and the 7.62-Mev gamma-ray lines are about the same. Crane, Delsasso, Fowler and Lauritsen,<sup>22</sup> in addition to the first five lines reported by Gaertner and Pardue, observed a gamma-ray line of 3.1-Mev energy. This could result from a transition between the 7.62- and the 4.37-Mev levels in  $\text{C}^{12}$ . Crane, Halpern and Oleson<sup>23</sup> report gamma-rays of energies 8.2, 6.6, 5.1, 4.1, and 2.5 Mev as coming from deuteron bombardment of nitrogen.

#### ACKNOWLEDGMENTS

The authors wish to express their appreciation of the continued encouragement and advice given them by Professors R. F. Bacher and H. A. Bethe. The  $\text{N}^{15}$  used in these experiments was kindly furnished by Professor H. C. Urey of Columbia University.

<sup>22</sup> H. R. Crane, L. A. Delsasso, W. A. Fowler and C. C. Lauritsen, Phys. Rev. **48**, 100 (1935).

<sup>23</sup> H. R. Crane, J. Halpern and N. L. Oleson, Phys. Rev. **57**, 13 (1940).