Transmutation Functions at High Bombarding Energies¹

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The differential transmutation functions of the reactions,

$$C^{12} + D^2 = N^{13} + n^1, \tag{1}$$

$$N^{14} + D^2 = O^{15} + n^1,$$
 (2)

$$O^{16} + D^2 = F^{17} + n^1, (3)$$

have been measured at bombarding energies from 2 to 5 Mev. All three curves showed a well marked change of slope at the top of the potential barriers. These points corresponded to potential barrier heights of 2.8, 3.2, and 3.1 Mev for carbon, nitrogen, and oxygen, respectively. These values are slightly more than half those reported for the same elements with alpha-particles. The absolute cross sections for disintegration at the top of the potential barrier are $4.0, 2.8, and 6.0 \times 10^{-26}$ cm² for the elements in order. At bombarding energies greater than the tops of the potential barriers the curves show interesting effects which are probably due to interfering side reactions.

INTRODUCTION

N a previous paper,3 work was described on the transmutation functions of carbon and oxygen up to bombarding energies of about three million volts. At that time it was noted that the transmutation function of carbon showed indications of flattening at the highest energies, and this was taken to indicate that the most energetic particles were approaching the top of the potential barrier of the carbon nucleus. When higher energies became available because of improvement of the cyclotron, this problem was reopened in order to study the shapes of the transmutation functions at energies near the tops of the potential barriers, and to study the behavior of the transmutation functions beyond the potential barriers. In order to reach more general conclusions, the three neighboring elements carbon, nitrogen, and oxygen were chosen for the study since they produce radioactive substances by the analogous reactions:

$$C^{12}+D^2=n^1+N^{13}$$
 ($\frac{1}{2}$ life=10.3 min.), (1)

$$N^{14} + D^2 = n^1 + O^{15} \left(\frac{1}{2} \text{ life} = 2.3 \text{ min.}\right), (2)$$

$$O^{16} + D^2 = n^1 + F^{17}$$
 ($\frac{1}{2}$ life = 1.2 min.). (3)

In this way it was hoped that one could detect special behavior of the transmutation functions due to any peculiarity of one of the nuclei. This was a fortunate precaution in as much as the three transmutation functions studied differed in shape very decidedly at bombarding energies greater than the heights of the potential barriers.

EXPERIMENTAL

The principles of the experimental method have been described previously. Briefly, it consists in gathering radioactive recoil atoms at various points along the path of a beam of deuterons as it is retarded while passing through a gas containing the element to be studied. In these experiments the recoil atoms were collected on platinum foils of about 1 cm stopping power, each of which were spaced about three millimeters apart. The platinum foils also served to cut down the range of the beam to a more convenient interval than the range of the beam in air (20 cm). The area of the foils was sufficiently greater than that of the beam so that recoil atoms, projected at an angle from the edge of the beam with a sufficient forward component of range to reach a foil, would strike the foil and not the frame on which it was supported. The spacing of the foils was slightly greater than the maximum forward range of the recoil atoms.

This method leads to a thin target transmu-

¹ A preliminary report on these experiments was given at the Berkeley Meeting, Nov. 28, 1935.

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³ Newson, Phys. Rev. 48, 790 (1935).

tation function since the target thickness is the average forward range of the recoil atoms which at the maximum energy was only about two millimeters. The transmutation functions are not subject to error due to variations in the intensity of the beam since all the foils are subject to the same fluctuations.

In the experiments, the bombarding chamber of the cyclotron was evacuated and then filled with nitrogen, oxygen, or carbon dioxide at atmospheric pressure. The bombardment was continued for a time greater than the half-life of the resulting radioactivity. The beam had an intensity of about two microamperes. The current was held as constant as possible in order to be able to calculate the absolute cross sections for disintegration. Ten collector foils were used in the experiment. After bombardment the decay of the activity on each foil was measured by measuring the activities of the foils in rotation. About four measurements were made on each foil so that a good decay curve was obtained for the desired activity; and, after the disappearance of this activity, any activity of longer half-life due to contamination could be measured and eliminated. The measurements were made on a Lauritsen type electroscope for which the fiber moved one division for every 10⁴ beta-rays which entered.

Corrections

The results of the measurements need to be corrected for two effects which vary with the energy of the deuteron:

First, the stopping power of the platinum foils varies decidedly within the range of energies which were used in this experiment. Corrections were made by using the calculations of Mano.⁴ It was necessary to use Mano's formula for energies at which it is not strictly valid. However, these points are below the potential barrier and consequently are of lesser interest in this experiment. Mano's formula has been tested recently by Laslet⁵ and found to be valid for deuterons of about the energies used. The stopping powers of the foils were measured originally with the alphaparticles of polonium. The correction of the stopping power found in this way amounted to

about twenty-five percent for the highest energies.

Second, the effective target thickness, which is the average forward range of the recoil atoms, varies with the deuteron energy. This quantity is

$$R_{t} = \frac{1}{2} \int_{0}^{\theta \max} (R \cos \theta) F(\theta) d\theta, \qquad (I)$$

where R is the range of the recoil atom, θ is the angle between the direction of the recoil atom and that of the bombarding particle, $F(\theta)$ is the distribution per unit angle of the recoil atoms, and θ max is the maximum value of θ , which may or may not be 180°. From the work of Blackett and Lees⁶ we may take the approximate expression

$$R = \frac{3}{4} V_r - 0.2, \tag{II}$$

where R is expressed in millimeters and V_r in 10^{-8} centimeters per second. Neglecting the constant term

$$R_t = \frac{3}{8} \int_0^{\theta \max} V_r \cos \theta F(\theta) d\theta.$$

This expression is more easily integrated in the coordinates of the center of gravity of the system. Let V be the velocity of the moving system, V' be the velocity of the recoil atom in the moving system, and ϕ be the angle between V and V'. Then, the forward component of velocity, $V_r \cos \theta = V + V' \cos \phi$, and, the distribution function, $F(\theta)d\theta = \sin \phi d\phi$ assuming that the distribution is isotropic in the moving system; the integration is to be taken over all values of ϕ . Making the above substitution:

$$R_{t} = \frac{3}{8} \int_{0}^{\pi} (V + V' \cos \phi) \sin \phi d\phi = \frac{3}{4} V.$$
(III)

Carrying out the integration without neglecting the constant term in Eq. (II) would make a negligible change in Eq. (III). In the case of nitrogen, recoil atoms are projected both forward and backward and the correction should take into account the average backward range which accounts for some of the activity. The correction has been worked out in this way, but the result does not differ essentially from Eq. (III). It is

⁴ Mano, J. de phys. 5, 628 (1934). ⁵ Laslet, Phys. Rev. 50, 391 (1936).

⁶ Blackett and Lees, Proc. Roy. Soc. 134, 658 (1932).



FIG. 1. Excitation curves of carbon, oxygen, and nitrogen. The ordinates are relative to the activity at the top of the potential barrier of each element. The ordinate 1 corresponds to the cross sections 4.0, 6.0, and 2.8×10^{-26} cm² for carbon, oxygen, and nitrogen, respectively.

apparent then, that the effective target thickness is proportional to the velocity of the deuteron, since

$$V = M_{\bullet D} V_D / (M_t + M_D),$$

where V_D and M_D are the velocity and mass, respectively, of the deuteron, and M_t is the mass of the bombarded nucleus.

The residual ranges of the deuterons at the front of each foil was converted into energy by the use of the Cavendish range-energy curve. Fig. 1 shows the transmutation functions in terms of the energy of the deuteron after making all corrections; the activity is plotted relative to the intensity at the top of the potential barrier. The absolute cross sections were calculated from the effective target thickness R_t from Eq. (III). The absolute cross sections at the tops of the potential barriers are 4.0, 2.8, and 6.0 cm²×10⁻²⁶ for carbon, nitrogen, and oxygen, respectively. These values are probably accurate within a

factor of two while their relative accuracy should be considerably better.

Results

All three curves show a very decided change of slope somewhat above three million volts. The almost discontinuous nature of the nitrogen and oxygen curves indicates that the tops of the potential barriers are very well marked. The energies at the points of inflection are 3.3 Mev, 3.7 Mev, and 3.5 Mev for carbon, nitrogen, and oxygen, respectively. Correcting for the motion of the nucleus, the energies at the tops of the potential barriers are 2.8 Mev, 3.2 Mev, and 3.1 Mev.

The value for the top of the potential barrier of nitrogen bombarded by alpha-particles is 5.6 Mev, and for carbon 5.1 Mev, according to Pollard.⁷

7 Pollard, Phys. Rev. 47, 611 (1935).

The top of the potential barrier may be considered to be the point at which the attractive force between the two nuclei just balances the Coulomb force of repulsion. If the attractive force, as is frequently assumed, starts at some critical value of the separation and increases extremely rapidly (having a potential curve of nearly infinite slope), the top of the potential barrier for both deuterons and alpha-particles would be located very near to this critical separation and the height for alpha-particles would be twice that for deuterons. It is seen from the above data that this picture is surprisingly close to the truth. However, the two sets of determinations should be repeated under similar experimental conditions before any very definite conclusions can be reached.

The behavior of the transmutation functions beyond the potential barrier is most erratic. The nitrogen curve comes nearest to what one would expect. According to the Gamow formula, the cross section for disintegration, $\sigma = k V^{-1} e^{-s}$, where V is the velocity of the bombarding particle, and s is a function of the bombarding energy. At the top of the potential barrier the exponential term should become constant, and σ should, therefore, be inversely proportional to the velocity. The small decrease beyond the potential barrier in nitrogen is probably due to this cause. There is, however, an interfering reaction which might cause this curve to decrease :

$$N^{14} + D^2 = C^{13} + He^3.$$
 (4)

It is impossible, at present, to investigate reaction (4) because the range of the He³ will always be considerably less than that of the bombarding deuteron.

In the previous paragraph the possibility of a transmutation being affected by side reactions is discussed. However, it is known from the work of Haxel⁸ that such interference between two concurrent reactions is possible. Haxel found that the transmutation function of the reaction

$$N^{14} + He^4 = F^{17} + n^1 \tag{5}$$

started with a threshold at 5 Mev; he observed at the same time that the transmutation function

of the concurrent reaction

$$N^{14} + He^4 = O^{17} + H^1$$
 (6)

dropped sharply at the same point where that of the first reaction started to rise. This showed very definitely the possibility of interference between two such reactions. This is most easily understood on the basis of the theory of the intermediate product put forward by Harkins⁹ and by Bohr.¹⁰ On this basis all the alpha-particles which enter the nitrogen nucleus are captured to form an unstable intermediate nucleus, F^{18} , which subsequently breaks up to give the final products of either reaction (5) or (6). Since the number of these intermediates is limited, the sudden increase of reaction (5) beyond its threshold must cause a corresponding decrease in reaction (6).

The slow rise in the transmutation function of oxygen (reaction(3)), after the top of the potential barrier has been reached, must be due to effects which are independent of the penetration of the deuterons into the nucleus. Similarly in the work of Haxel, reaction (5) has a threshold at a bombarding energy near the top of the potential barrier, but, in spite of this, the transmutation function rises with the bombarding energy. Reaction (3) starts from a threshold at 2 Mev so that it is not surprising that its excitation curve should continue to rise beyond the potential barrier.

The sharp drop in the transmutation function of carbon (reaction (1)) is probably due to the reaction:

$$C^{12} + D^2 = B^{10} + He^4.$$
 (7)

The transmutation function of this reaction must rise rapidly beyond the potential barrier to cause the decided drop in the intensity of reaction (1). Unfortunately, reaction (7) like reaction (4) cannot be studied directly.

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

⁸ Haxel, Zeits. f. Physik 93, 400 (1935).

⁹ Harkins, Phys. Rev. **44**, 530 (1933); Science **83**, 533 (1936); Phys. Rev. **51**, 52 (1937); Proc. Nat. Acad. Sci. **23**, 120 (1937).

¹⁰ Bohr, Nature **137**, 344 (1936).