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Experiments with High Velocity Positive Ions. III.—The Disintegration of Lithium, Boron, and Carbon by Heavy Hydrogen Ions.

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1. Introduction.

An important enlargement of the field of nuclear research has been effected by the experiments of Lawrence, Lewis, and Livingstone,* who first used the ions of the heavy isotope of hydrogen to produce nuclear disintegrations. Their pioneer experiments showed that these ions, which will in future be termed "diplons,"† produced disintegrations of considerably greater complexity than those produced by protons of the same energies. Thus, Lawrence, Lewis, and Livingstone reported that from most of the substances bombarded, one or more groups of protons were emitted, whilst a group having a range of about 18 cm. appeared to be emitted from every element investigated. This surprising result led these workers to the hypothesis that the "diplon" is unstable in a strong nuclear field and that it breaks up into a proton and a neutron with a liberation of energy of the order of 5 million volts, the neutron mass being taken to be 1.0006; to explain the observed proton energies.

In addition to the emission of long-range protons, Lawrence, Lewis, and Livingstone (*loc. cit.*) reported the emission of α -particles from lithium, beryllium, boron, nitrogen, magnesium, and aluminium. The experiments of Rutherford, Oliphant, and Kinsey§ and the Wilson chamber photographs of Dee and Walton|| showed that the 13.2 cm. α -particles from lithium originated from the disintegration of Li⁶ into two α -particles, and suggested that Li⁷ disintegrated into two α -particles and a neutron. No detailed studies have been made of the disintegration of the other elements.

The experiments described below have been made with a view to obtaining more evidence on the nature of the disintegrations in which protons are emitted; we have also studied the emission of α -particles from boron under diplon bombardment.

* ' Phys. Rev.,' vol. 44, p. 55 (1933).

† ' Proc. Roy. Soc.,' A, vol. 144, p. 1 (1934).

‡ ' Phys. Rev.,' vol. 44, p. 778 (1933).

§ 'Proc. Roy. Soc.,' A, vol. 141, p. 722 (1933).

|| ' Proc. Roy. Soc.,' A, vol. 141, p. 733 (1933).

2. Apparatus.

In these experiments, we have used the high voltage D.C. generator described in our previous paper,* which gives a maximum steady potential of 700 kv. The tube for the acceleration of ions there described has been modified to give larger intensities of ions and more reliable operation. Porcelain cylinders of 12-inch diameter and 3 feet 6 inches length have been substituted for the glass cylinders used previously; a discharge tube of the type described by Rutherford and Oliphant' has been transferred from the apparatus of Dee and Walton to the new apparatus and the evacuation of the tube is now carried out by a Metropolitan-Vickers 04 Apiezon Oil Diffusion Pump which has a speed of 1200 litres/sec. for hydrogen. This great increase in pumping speed has increased considerably the reliability of operation of the tube; the new discharge tube enables total currents of 200 microamperes to be obtained, whilst, in spite of using double stage acceleration, the geometry of the electrodes is now such as to focus about 20 per cent. of the ions into a beam having a diameter of only 1 cm. in the experimental chamber, thereby increasing considerably the fraction of disintegration particles which can be made to enter the recording apparatus.

3. The Detection of High Speed Protons.

In most experiments we have used the scintillation method for fixing approximately the ranges of the particles emitted, carrying out a detailed study with the ionization chamber, linear amplifier, and oscillograph previously described. In interpreting records of proton emission, difficulty arises when the background disturbance of the oscillograph record due to electrical and acoustical "pickup" is comparable with the oscillograph deflection due to the protons. The greatest care has been taken to reduce as far as possible the magnitude of this pick up, but in practice there is often a disturbance due to the discharge tube or to soft radiation which gives a background oscillation on the oscillograph. Consequently it is often necessary to count only deflections above some arbitrary size corresponding to protons approaching the end of their range. The absorption curves obtained are thus of a differential type, maxima being obtained near the end of the range of a group of protons.

In the present experiments we have not applied magnetic analysis to separate out the protons and diplons in our beams. Since we usually have a mixture of hydrogen and diplogen in our discharge tube, the beam of ions contains

^{* &#}x27; Proc. Roy. Soc.,' A, vol. 136, p. 619 (1932).

^{† &#}x27; Proc. Roy. Soc.,' A, vol. 141, p. 259 (1933).

protons of full and half energy and diplons of full and two-thirds energy. This spread of the energy causes the peaks in the absorption curves to be less sharply defined, but does not prevent an accurate deduction of the range from being made.

In most of these experiments, an ionization chamber having a depth of 1 cm. has been used; in general a correction of 0.5 cm. has been made to the ranges obtained from the absorption curves to allow for this depth.

4. The Disintegration of Lithium.

A target of pure Kahlbaum lithium, which on exposure becomes covered with a layer of lithium hydroxide, was bombarded with a mixed beam of ions obtained from hydrogen containing approximately 10 per cent. of diplogen. A target of 5 cm. diameter was used, the beam covering a diameter of about 1 cm.; the distance from centre of target to recorder was 5 cm., and the solid angle subtended by the counter aperture at the recorder was about 1/30. With this arrangement, the emission of particles having a range of about 30 cm. was observed, and the absorption curve of fig. 1 was obtained for an accelerating



potential of 500 kv. The maximum number of particles counted was about 700 per minute. The observations show that a group of protons having a range of $30.5 \text{ cm.} \pm 1 \text{ cm.}$ for an accelerating potential of 500 kv. is emitted from lithium. Observations were not carried out at absorptions less than 14 cm. owing to the presence of the strong α -particle group of 13.2 cm. range. The range observed is consistent with the observation of Lawrence, Lewis, and Livingstone that protons of range up to 40 cm. were emitted from lithium fluoride when bombarded with their more energetic particles.

5. The Mechanism of the Disintegration.

If we make the assumption that the following nuclear reaction occurs

$$_{3}\mathrm{Li}^{6} + {}_{1}\mathrm{D}^{2} = {}_{3}\mathrm{Li}^{7} + {}_{1}\mathrm{H}^{1}$$

the kinetic energy change in the transmutation may be calculated from the mass change

$$(6 \cdot 0145 + 2 \cdot 0136) - (7 \cdot 0146 + 1 \cdot 0078) = 0 \cdot 0057 \pm 0 \cdot 0007 = 5 \cdot 3 \pm 0 \cdot 6 \text{ million volts.}$$

Allowing for the kinetic energy of the diplons and the energy taken by the recoiling nucleus, the disintegration protons may be expected to have an energy of $5 \cdot 0 \pm 0.5$ million volts. If we adopt Duncanson's figures* for the range velocity function of the protons, protons of this energy would have a range 35 ± 5 cm. The observed ranges would thus be consistent with the hypothesis as to the nature of the disintegration. If, on the other hand, we take the masses of Li⁶ and Li⁷ to be fixed by disintegration data, the expected energy in the protons is $5 \cdot 1 \pm 0 \cdot 2$ million volts, which, from Duncanson's data would give a range of 35 ± 2 cm. The agreement while fair is not as good as might be expected; the discrepancy may be due to the uncertainties in the range velocity relation for protons, to the emission of radiation in the transmutation, or it may be possible that the unstable nucleus Be⁸ may loose part of the energy imparted by the diplon before breaking up.

Strong support for the proposed reaction is given by the experiments of Oliphant, Shire, and Crowther[†] on the transmutation of the separated isotopes of lithium, which showed that the 30 cm. protons result from the bombardment of Li^6 by H².

6. The Number of Disintegrations.

The absolute number of disintegrations cannot be obtained with any precision from our present experiments owing to the uncertainty in the composition of the beam. For the same reason, no accurate determination of the variation of the number of disintegrations with voltage has been made. The results for the mixed beam are, however, given in fig. 2. It is seen that there is no sharply defined lower limit to the voltage at which disintegrations can be observed, the curve being similar to that obtained for the $13 \cdot 2$ cm. α -particles emitted in the disintegration

$_{3}\mathrm{Li}^{6} + _{1}\mathrm{D}^{2} \rightarrow 2_{2}\mathrm{He}^{4}.$

* 'Proc. Camb. Phil. Soc.,' vol. 30, p. 102 (1934).
† 'Nature,' vol. 133, p. 377, March 10 (1934).

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We find that the number of 13.2 cm. α -particles emitted is roughly twice the number of disintegration protons for diplon energies of 500 kv., so that the relative probability of the two types of disintegration is not very different.

7. The Disintegration of Carbon.

The absorption curves obtained for a target of Acheson graphite with the same experimental arrangement as for lithium are given in fig. 3, the maximum number of disintegration particles being of the same order as that found for



lithium. The curves suggest the presence of a group of particles having a range of $14 \cdot 0 \pm 1$ cm. for an accelerating potential of 500 kv. An analysis of the oscillograph deflections suggests that this group are protons, and it also indicates the presence of a weak proton group having a range of about 8 cm.

Taking the energy of the 14 cm. protons as $2 \cdot 9$ million volts, the kinetic energy change in the reaction is $2 \cdot 6 \pm 0 \cdot 16$ million volts.

The range of the proton group obtained is about 1 cm. longer than would be expected from the observations of Lawrence, namely, that, using $1 \cdot 2$ million volt diplons, the proton range is 18 cm.

Since these experiments were completed, Lauritsen and Crane* have reported the emission of a γ -ray having an energy of about 3 million volts when carbon

* ' Phys. Rev.,' vol. 45, p. 345 (1934).

is bombarded with diplons. If we assume that the 14 cm. proton and the γ -ray are emitted at the same time from the reaction

$$_{6}C^{12} + _{1}D^{2} = _{6}C^{13} + _{1}H^{1} + h\nu$$

we obtain the following value for the mass of C^{13} :

$$12 \cdot 0036 + 2 \cdot 0136 - 1 \cdot 0078 - 0 \cdot 0027 - 0 \cdot 0032 = 13 \cdot 0035 \pm 0 \cdot 0006.$$

The band spectrum mass for C¹³ is 13.0039 ± 0.0013 . The mass obtained from the disintegration of B¹⁰ by α -particles is 13.0047 ± 0.0005 (Chadwick, Constable, and Pollard).*

If the reaction assumed is correct, it might be expected that some transmutations would occur in which no γ -ray was emitted, the full energy appearing in the form of kinetic energy, so that protons of about 40 cm. range should be



observed. It appears certain, however, that if they exist the number of such protons must be less than 1 in 500 of the number of 14 cm. protons.

On the other hand, the emission of the γ -ray observed by Lauritsen and Crane may be connected with the transformation of C¹² to N¹³ under proton bombardment with the subsequent emission of a positive electron.[†] If this is so, then the mass of C¹³ calculated from both reactions would be about 13.006.

Evidence to be referred to later suggests that the 8 cm. proton group may be due to a layer of adsorbed oxygen.

Fig. 4 gives the observed variation in the number of disintegrations with • voltage. These results were obtained from a beam of mixed ions, and an absolute determination of numbers has not yet been made.

* ' Proc. Roy. Soc.,' A, vol. 130, p. 463 (1933).

† Cockcroft, Gilbert, and Walton, 'Nature,' vol. 133, p. 328 (1934).

8. The Disintegration of Boron.

A study of the particles emitted from a target of boron under diplon bombardment has shown that very swift α -particles are emitted together with at least three groups of protons. The absorption curve is given in fig. 5.

By combining deductions from the shape of the absorption curve, fig. 5, with an analysis of the distribution of the size of oscillograph deflections we find that both α -particle and proton groups are present. A complete interpretation



FIG. 4.-Diplons on carbon target ; absorber 8.9 cm. of air.

of these curves up to 16 cm. is difficult owing to the probable superposition of one or more proton groups on a continuous distribution of α -particles. The existence of the following groups of particles seems, however, to be certain :—

- (1) α -particles of all ranges up to a maximum of 15.0 ± 1 cm.
- (2) A proton group of range 31 ± 2 cm.
- (3) A proton group of range 58 ± 2 cm.
- (4) A proton group of range 92 ± 2 cm.

The rise in the absorption curve from 6 to 10 cm. suggests the presence of another proton group, but until Wilson chamber photographs are available it is not possible to be certain of the interpretation.

The following tentative explanations are advanced for groups (1) to (4).

(1) The continuous distribution of α -particles corresponds to the disintegration of B¹⁰ into three α -particles

$$_5\mathrm{B^{10}} + _1\mathrm{D^2} \rightarrow 3_2\mathrm{He^4}.$$

The absorption curve for this disintegration would be expected to be of a similar nature to that observed for the disintegration of B¹¹ by protons.* If



FIG. 5.—a-particles and protons from boron. Diplon energy 500 kv.

•the hump in the curve at 9 cm. is due to a superposed homogeneous group of protons, then the residual curve would have a shape similar to the B¹¹ curve.

If the end-point of the α -particle curve corresponds to a disintegration in which one α -particle is ejected in a direction opposite to the other two, then

* Rutherford and Oliphant, ' Proc. Roy. Soc.,' A, vol. 141, p. 259 (1933).

this α -particle possesses two-thirds of the total kinetic energy. The total kinetic energy in the disintegration particles would thus be

 $(12 \cdot 3 \pm 0 \cdot 30) \times 3/2$ million volts = $18 \cdot 45 \pm 0.45$ million volts.

If the mass of B¹⁰ be taken as $10 \cdot 0135 \pm 0 \cdot 0005$ the expected kinetic energy is $19 \cdot 4 \pm 0 \cdot 6$ million volts. The agreement is satisfactory, but it is very desirable to have further evidence about this disintegration from Wilson chamber photographs. It should be noted that some of the α -particles from this reaction are swifter than any which have previously been discovered. The fastest α -particles from radioactive bodies have a range of $11 \cdot 5$ cm. whilst the α -particles discovered by Lawrence in the reaction Li⁶ + D had a range of 14 cm.

(2), (3), and (4) assuming Duncanson's range velocity relation for protons, these groups correspond to protons of energy 4.7, 6.7, and 8.8 million volts, and could be explained by the following reaction:

$$_{5}B^{10} + _{1}D^{2} = _{5}B^{11} + _{1}H^{1} + hv.$$

If the proton of range 92 cm. is emitted when all the energy of the reaction goes into kinetic energy, we may calculate the difference of mass of B^{10} and B^{11} as follows:

 $B^{11} - B^{10} = 2 \cdot 0136 - 1 \cdot 0078 + 0 \cdot 0005 \text{ (K.E. of } D^2\text{)}$ $- 0 \cdot 0102 \text{ (K.E. after disintegration)}$ $= 0 \cdot 9961 + 0 \cdot 0004.$

The difference of mass from Aston's determination is $11 \cdot 0110 - 10 \cdot 0135 = 0 \cdot 9975 \pm 0 \cdot 0007$. Thus the agreement is not very good. If, on the other hand, we were to assume that the masses of B¹⁰ and B¹¹ could be obtained from the end-point of the continuous α -particle distributions in the transmutations by diplons and protons respectively, we should obtain very close agreement with these experiments.

The two groups of lower energy may correspond to disintegrations in which energy of 2 and 4 million volts respectively is emitted in the form of γ -radiation.

The total number of particles in groups (2), (3), and (4) is approximately one-third of the maximum number of α -particles observed. This suggests that the disintegration leading to three α -particles has about the same probability as the disintegration leading to B¹¹.

9. The Emission of Particles on the Bombardment of Heavy Elements by Diplons.

A detailed study has been made of the particles emitted from copper, copper oxide, iron, iron oxide, tungsten, and silver. In order to facilitate comparison of these elements the target illustrated by fig. 6 was constructed so that four targets could be interchanged by rotation of a ground joint. With this arrangement the solid angle subtended by the window at the target was increased to 0.25; a still further increase in the number of particles observed was obtained by using a helium-diplogen mixture containing 30% of diplogen. A series of eight experiments was carried out, using diplon energies of 500 kv.

(i) and (ii) The emission of particles from carbon, copper oxide, copper, and iron was compared in one series of experiments and of tungsten, silver, iron oxide, and iron in another, the absorption curves of figs. 7, a, and 7, b, being obtained. Observations could not be carried out on silver at less absorber than 6 cm. owing to the emission of soft radiation in considerable intensity.

From these curves and from an analysis of the size of the oscillograph deflections we conclude—

(a) that a group of protons of range about 14 cm. is emitted from all these targets, the numbers from carbon being about ten times the numbers from other targets;



- (b) that a group of protons of range 8 cm. is emitted from all the targets;
- (c) that a group of singly charged particles of range about 4 cm. is common to all targets except carbon;
- (d) that α -particles are emitted from all the targets, the numbers falling rapidly for absorbers greater than 2 cm., a few particles being observed up to 5 cm.
- (e) that whilst the 8 cm. group is stronger from Fe_2O_3 than from Fe, the relative strength of the 14 cm. groups are reversed.

(iii) A strip of the tungsten used in experiment (ii) was washed in succession in caustic soda, distilled water, ether, caustic soda, and distilled water and

then bombarded. The absorption curve of fig. 8, b, was then obtained, showing that whereas the short range group of particles and the number of particles at 8 cm. absorber had roughly the same relative magnitude as in experiment (ii) the 14 cm. peak was reduced in numbers to about one-third of the 8 cm.





(iv) At the suggestion of Dr. Oliphant the target was heated in position to about 2000° C. by passing a heavy current through it, and the emission of particles observed under these conditions and immediately after heating. The results showed a considerable diminution in the number of particles in the 8 cm. groups and short-range groups, whilst the numbers in the 14 cm.

group were reduced by a further factor of 2, the curve obtained after heating being shown on fig. 8, c.

(v) The target was left for 18 hours in the apparatus and then bombarded whilst cold. The number of particles observed was if anything smaller than immediately after heating the target, showing that no new layers of impurities had been adsorbed during this period.



(vi) Carbon dioxide was admitted to the apparatus to a pressure of 1 cm., the target flashed at about 1600° C. and again bombarded. No change in the number of particles was observed from experiment (v), showing that no surface layer had been adsorbed in sufficient amount to give rise to an appreciable effect. Downloaded from http://rspa.royalsocietypublishing.org/ on July 7, 2018

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(vii) After being cleaned by heating, the target was taken out and oxidized by passing an electric current through it, replaced and again bombarded. The absorption curve obtained is shown in fig. 8, d. The short-range group and





8 cm. group have again appeared strongly together with a few particles at absorptions of 14 cm. about equal in numbers to those obtained in (iii).

(viii) The target was cleaned by heating in vacuum, was removed and oxidized and then handled in order to reproduce the treatment which a metal

might reasonably receive before mounting in an apparatus. The target was then replaced and bombarded and the curves of fig. 8, e, were obtained. We see that all three peaks now appear strongly; the long-range peak is again stronger than the 8 cm. peak and is now restored to its former position.

These experiments may be summarized as follows :---

(i) The 8 cm. proton group, the 4 cm. proton group, and the short-range α -particle groups disappear when the target is heated strongly and reappear when the target is oxidized. The evidence suggests therefore that these groups are due to oxygen.

Other experiments carried out by Rutherford and Oliphant* have lead to similar conclusions and they propose the following explanation for the reaction leading to the 8 cm. protons.

$_{8}O^{16} + _{1}D^{2} = _{8}O^{17} + _{1}H^{1}$

the observed range being in good accord with the accepted masses for O^{16} and O^{17} . The explanation of the 4 cm. proton group must await further experiments; the energy difference between the two groups is 0.75 million volts.

(ii) The long-range proton group from the heavy elements appears to be complex, the greater part of the effect being removed by chemical cleaning of the surfaces, a small residual effect remaining due to protons having a slightly longer range. The coincidence of the range of the majority of the protons with the long-range group from carbon suggests therefore that a thin film of grease or other carbon impurity is responsible for the greater part of the effects.

An alternative explanation seemed, however, to be offered by the experiments of Rutherford, Harteck, and Oliphant,[†] which showed that on bombarding by diplons, ammonium sulphate, in which the ordinary hydrogen had been largely replaced by diplogen, very great numbers of particles were emitted, half of the particles having a range of 1.7 cm. and half a range of about 14 cm. It seemed, therefore, possible that the whole of our results might be explained by diplogen being absorbed on carbon from the bombarding stream, the adsorbed layer being then disintegrated by the diplon stream.

In order to test this hypothesis, we have carefully compared the ranges of the proton groups emitted from $(ND_4)_2SO_4$ and carbon under identical conditions. If all the results are due to adsorbed diplogen, we ought to get the

* In course of publication.

† 'Nature,' vol. 133, p. 413, March 17 (1934).

same 14 cm. range from the two substances, and we ought to observe large numbers of singly charged particles having a range of about 2 cm. from carbon.

The results of these experiments are given by figs. 9 and 10. In fig. 9 the absorption curves for the two substances are given, showing that the range of the protons from $(ND_4)_2SO_4$ is greater by about 1 cm. than the range of the protons from carbon. A still more conclusive proof of the difference between the two reactions is given by fig. 10 in which we plot the distribution in size of the oscillograph deflections for the two substances for small absorptions. Remembering that a large deflection corresponds to a proton near the end of



FIG. 9.—Comparison of ranges of long-range protons from $(ND_4)_2SO_4$ and C. D accelerating voltage of diplons 150 kv.; proton range 14 \pm 0.5 cm. C accelerating voltage of diplons 370 kv.; proton range 12.5 \pm 0.5 cm.

its range, we see at once that there are very few short-range particles from carbon compared with the number from $(ND_4)_2SO_4$. Experiments with greater thickness of absorber showed that the number of 14 cm. protons are of the same order of magnitude for the two targets.

We conclude that-

- most of the 14 cm. protons emitted from carbon are due to the transmutation of C¹² into C¹³;
- (2) there is probably a small additional group of protons of range 15 cm. at 500 kv. due to the adsorption of diplogen and the subsequent disintegration of the adsorbed layer;

- (3) the greater part of the effects observed with the heavy elements is due to thin films of grease or other form of carbon impurity adsorbed on the surface;
- (4) a smaller number of 15 cm. protons are emitted from the heavy elements due to the adsorption of diplogen from the bombarding stream.

10. Conclusions.

We may summarize the experimental results by stating (i) that the emission of long-range protons is in all substances investigated (D, Li, B, C, O) bound up



(a) and (a') 0.63 cm. absorber.
(b) and (b') 1.23 cm. absorber.

with the transformation of an element into the next heavier isotope of that element, the neutron being captured and the proton ejected; (ii) that when transmutations leading to the emission of an α -particle are possible (Li, B) the probability of this transmutation is of the same order as that of the first type; (iii) that the emission of protons from the heavier elements (Cu, Fe, Ag, W) is in our experiments due to films of impurities of oxygen, carbon, and adsorbed diplogen.

⁽c) and (c') 2 cm. absorber.

We wish finally to express our appreciation of Lord Rutherford's constant interest and encouragement, and to thank Dr. Harteck for his work in the preparation of the diplogen. We are indebted to the Government Grant Committee of the Royal Society for a grant in aid of the purchase of the fast pumping equipment used; one of us (E. T. S. W.) wishes to acknowledge the grant of a Senior Research Award of the Department of Scientific and Industrial Research.

Summary.

The disintegration of lithium, boron, and carbon, when bombarded by fast ions of heavy hydrogen, has been examined. For lithium, a group of protons of 30.5 cm. range has been observed; this can be interpreted as resulting from the transformation of Li⁶ to Li⁷. Boron gives proton groups of ranges 92 cm., 58 cm., and 31 cm. together with a continuous distribution of α -particles with ranges up to a maximum of 15 cm. Carbon gives a group of protons with a range of 14 cm. These would correspond with the 18 cm. group observed by Lawrence from all targets if allowance is made for the higher energies of his bombarding particles.

A 14 cm. group of protons has been observed from a number of heavy elements. Experiments have been carried out which indicate that these are probably due to carbon present on the surface in a thin film of grease. A smaller number of protons having a slightly longer range are also emitted, these are probably produced from the disintegration of diplogen adsorbed on the surface. Layers of oxygen on the surface may also give rise to spurious effects.

The emission of a fast group of protons always appears to be connected with the change of one isotope of an element into its next heavier isotope.