The Transmutation of Fluorine by Proton Bombardment and the Mass of Fluorine 19

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Fluorine has been transmuted by bombardment with protons into oxygen and helium. The number of transmutations has been followed from a proton energy of 0.675 to 1.63×10^6 electron-volts. The alpha-particles emitted have a range greater than 6 cm. Their range increases with proton energy in the expected manner. The short range alpha-particles observed both by us and by other workers are probably due to a boron contamination. Professor Oppenheimer has calculated, along the lines of the Gamow theory, the manner in which the probability of transmutation should vary with the energy of the protons. The measurements are in excellent agreement with the theory. The mass of the fluorine atom that is calculated from the reaction, 19.0031, is in excellent agreement with other transmutation data, but not with the value usually accepted, namely 19.000.

Introduction

Using 450,000 volt protons in their pioneer experiments on the artificial transmutation of the elements, Cockcroft and Walton[1] observed the emission of 3.0 cm alpha-particles from a target of CaF_2. They attributed this emission to the transmutation of fluorine with the formation of oxygen. Later Oliphant and Rutherford,[2] in careful experiments with much greater numbers of protons at lower voltages, observed 4.1 cm alpha-particles from a target of FeF_2, which also were attributed to fluorine. In our preliminary experiments[3] using protons of much greater energy, we observed the emission from CaF_2, in addition to the particles of short range, of alpha-particles of about 7 cm range. We have in the interim carried out a more thorough experimental investigation and have come to the conclusion that the long range group alone is emitted by fluorine,[4] and that the particles of shorter range were the result of a contamination, probably by boron.

Method and Apparatus

The apparatus used in this work was the larger of the two that have been developed in this laboratory for the production of high speed light ions without the use of high voltages. It has already been described[5] in this journal. The smaller apparatus has been used for work with lithium[6] and boron.[7]

The apparatus is so constructed that only ions having a particular value of e/m are accelerated. Heretofore we have used the hydrogen molecular ion, $(H)_2{}^+$. The disadvantage inherent in using this ion is that the deuton, $(H^2)^+$, has almost exactly the same e/m, and, if present in the apparatus, is accelerated at the same time. Since the deuton has proved to be a very reactive projectile in transmutation work and since it is always present in our apparatus in considerable numbers, the apparatus was adjusted for the present experiments to accelerate protons, $(H)^+$, only. Proton beams of energies up to 1.63 MEV (million electron-volts) were used.[8] Although currents of as much as 0.8 microampere were obtainable, generally much smaller currents than this were used because the yield of transmutations is very large at high voltages.

The energy of the bombarding protons was determined, whenever possible, by measuring their range when scattered into the ionization chamber by a platinum target. To obtain energies different from the one for which the apparatus was adjusted, mica foils of known stopping power were inserted in the path of the

[1] J. D. Cockcroft and E. T. S. Walton, Proc. Roy. Soc. **A137**, 229 (1932).

[2] M. L. E. Oliphant and Lord Rutherford, Proc. Roy. Soc. **A141**, 259 (1933).

[3] E. O. Lawrence and M. S. Livingston, Phys. Rev. **44**, 316A (1933).

[4] We have just seen the abstract of a paper by M. A. Tuve, L. R. Hafstad and O. Dahl presented at the April, 1934, meeting of the American Physical Society, in which they report finding 60 mm alpha-particles from fluorine, in agreement with our results.

[5] E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 608–612 (1934).

[6] M. C. Henderson, Phys. Rev. **43**, 98 (1933).

[7] M. G. White and E. O. Lawrence, Phys. Rev. **43**, 304 (1933).

[8] We have included in the results two runs in which the molecular ion beam was used. The deutons present do not seem to cause spurious effects with this particular target.

primary beam—thus the lowest value of the energy was obtained by inserting a 21 mm air-equivalent mica foil in the path of a 1.2 MEV beam.

The target was a piece of calcium fluoride melted onto a piece of platinum mesh and placed on the target-mounting in the Faraday chamber. It was one of twelve targets, any one of which could be rotated into the path of the proton beam by turning a ground joint. The targets included nearly all the elements in the first two rows of the periodic table and calcium hydroxide as well. Only from the calcium fluoride target was there any evidence of a 6 to 7 cm alpha-particle. As no alpha-particles of this range could be detected from any other target at the highest currents obtainable, they are certainly to be attributed to the transmutation of fluorine.

The alpha-particles were detected in a shallow ionization chamber by a four stage amplifier[9] of the type described by Wynn-Williams.[10] The Geiger counter used in earlier work was discarded for obvious reasons. The plate circuit of the third stage of the amplifier operated two separate output tubes. One of them actuated a cathode-ray oscillograph and the other a "scale of eight" thyratron counter.[11] The cathode-ray tube was a new type having a brilliant spot that is easily visible in daylight. It was kept under continuous visual observation during a run. We were thus able to keep close track both of all radiations that entered the ionization chamber and of the performance of the amplifier.

Visual observations with a cathode-ray oscillograph (combined with a counting device) have a twofold advantage. On account of the very long time scale, about two inches per sixtieth of a second, spurious impulses that are recorded by the counter are readily distinguished from the characteristically shaped potential pulses caused by alpha-particles. Also, with the oscillograph time scale oscillating in synchronism with the 60 cycle alternating potential that is applied to the plate of the main oscillator, it is possible to observe whether any pulses occur after the proton bombardment ceases. (The proton bombardment is confined to about 50 percent of the positive half of each cycle, or about 1/4 of the whole.) It should thus be possible to detect such effects as radioactivity of very short life. All alpha-particles or disintegration protons thus far observed have appeared simultaneously with the proton beam. None have been observed later than 1/300 sec. after its disappearance.

Range of the Alpha-Particle

Fig. 1 is a number-range curve for the alpha-particles from a target of calcium fluoride. The two scale of ordinates are given in absolute terms—that is, they represent the numbers of transmutations per 10^9 protons incident on the target. In calculating numbers of transmutations from numbers of alpha-particles it is assumed that the alpha-particles are emitted isotropically. Curves I, II and III are plotted on the 0 to 10 scale, the remainder on the 0 to 100 scale.

For comparison the number-range curves of alpha-particles from boron and lithium are given, curves VII and VIII. The extrapolated range of the lithium and boron curves are larger than the values given by other workers[1,2]

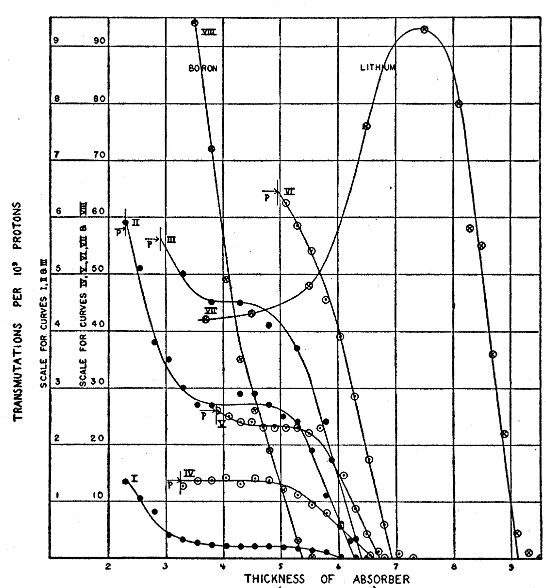

Fig. 1. Number of transmutations per 10^9 protons *vs.* absorber thickness for different proton energies.

[9] The amplifier and recording mechanism are to be described in more detail elsewhere.

[10] C. E. Wynn-Williams and Ward, Proc. Roy. Soc. **A132**, 391 (1931).

[11] C. E. Wynn-Williams, Proc. Roy. Soc. **A136**, 312 (1932).

because the energy of the bombarding proton is considerably greater. The increase in range agrees well with the amount calculated from the increase in proton energy. The difference in range between the boron or lithium alpha-particles and the fluorine alpha-particles should be more accurate than the actual measured range of either.

In calculating the curves for the three different elements, account has been taken of the fact that the mechanism of transmutation is different in each case. Thus, for fluorine one alpha-particle is emitted per transmutation. For lithium, however, there are twice as many, and for boron, three times as many alpha-particles as transmutations.

Curves I to VI were obtained when fluorine was bombarded by protons of the following energy: Curve I—0.675 MEV, II—0.975, III—1.10, IV—1.20, V—1.33, VI—1.63. Curve VII is lithium bombarded by 1.5 MEV protons, and Curve VIII boron bombarded by 1.2 MEV protons.

From the six curves of fluorine alpha-particles it can readily be seen that the range of the alpha-particle increases with the energy of the proton that produces it. From the mechanism of the transmutation it may be inferred that the alpha-particle receives approximately 4/5 of the energy of the proton and the recoiling nucleus 1/5. This conclusion is in excellent agreement with the data.

The curves are broken off at the left either at 2.3 cm, which was the least thickness of absorber possible, or else at the point where scattered primary protons began to enter the ionization chamber. There are so many more scattered protons than there are alpha-particles that they completely obscure the record if they enter the chamber. Some sort of differential ionization chamber will be necessary in further work with protons of this or higher energy.

The increased heights in the curves which come as the absorber thickness is reduced below about 3 cm, indicating alpha-particles of shorter ranges, are caused by alpha-particles from some contamination common to all targets. Further work definitely showed this contamination to be boron. Compared to other substances the yield of alpha-particles from boron is enormous. Each disintegration produces three alpha-particles and the number of disintegrations itself is about ten per million protons at a million volts. The peculiar form of the boron range curve makes contamination with boron particularly treacherous, as has already been emphasized by Oliphant and Rutherford[2] and by Tuve.[12] The exponential increase in number of alpha-particles makes it easy to determine a false alpha-particle range for an element that, in point of fact, yields no alpha-particle. Any alpha-particle with range less than four cm is suspect, unless special precautions are taken to ensure that no boron is present. The alpha-particles of short range that we detected from a number of targets in our early work are almost certainly the result of boron contamination.[3]

On the other hand, there is a very definite end to the boron range curve, as may be seen from Fig. 1, Curve VIII, or from Oliphant and Rutherford,[2] p. 270. The alpha-particle yielded by a calcium fluoride target goes at least a centimeter further than the boron alpha of longest range.

Voltage Transmutation Function

Fig. 2 shows the voltage transmutation functions of two targets: calcium fluoride and lithium. The lithium target was originally a piece of metallic lithium, but by the time the measurements were taken its surface had probably become lithium hydroxide, or even lithium carbonate. The ordinates in this figure are the heights of the plateaux of the number-range curves in Fig. 1. The plateau for Curve VI was estimated in an obvious manner by comparison with the others.

In this figure the number of transmutations is plotted against the range of the protons producing the transmutation and not against their energy directly. There is given a second scale of abscissas showing the proton energy in kilovolts to which the proton ranges correspond. This method of plotting shows at once in the case of lithium where the initial exponential rise with voltage ceases, and the increased number of transmutations becomes a result simply of the

[12] Tuve, Hafstad and Dahl, Phys. Rev. **43**, 942L (1933).

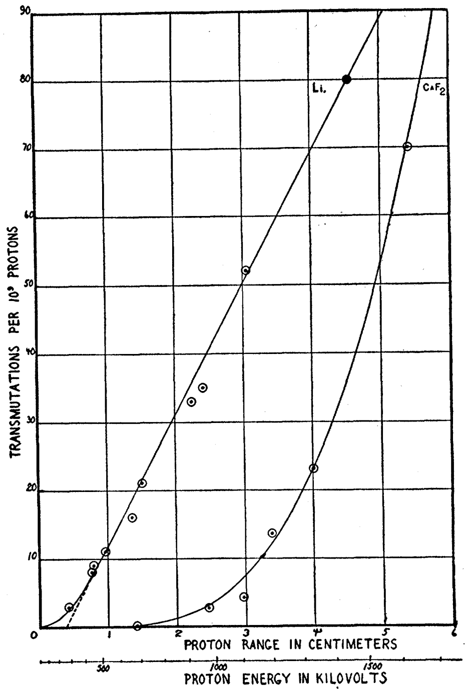

FIG. 2. Transmutations per 10^9 protons *vs.* proton range (energy) for Li and CaF_2.

increased penetration of protons into the target. The relation between proton range and proton energy was taken from data given by Blackett.

The lithium curve was taken from the work of Henderson.[6] The highest point (filled circle), however, was determined separately in the larger apparatus in the course of the present experiments.

We are greatly indebted to Professor Oppenheimer for working out, along the lines of the Gamow theory* of the nucleus, a theoretical formula for the voltage transmutation function of fluorine. Approximations that were sufficient in the case of lithium[6] are no longer so for fluorine. The assumptions underlying the theoretical formula are: (1) the peak in the potential barrier is high compared to the proton energy; and (2) the barrier is nearly impenetrable. The probability of transmutation is the product of two exponentials, corresponding to the two terms in the brackets, one representing the probability of a proton penetrating the barrier and the other the probability of transmutation after penetration. The fact that the target is "thick" also is taken into account. The formula is

$$N = kVe^{-S}(1+V^{\frac{1}{2}}/6+V/24+V^{\frac{3}{2}}/72+\cdots)$$

where

$$S=(4\pi^2e^2/h)\{Z_F[M_P/2E_p]^{\frac{1}{2}} + Z_\alpha Z_0[M_\alpha \cdot M_0/2(M_\alpha+M_0)(E_d+E_p^*)]^{\frac{1}{2}}\}.$$

Z_F, Z_0 and Z_α are, respectively, the atomic numbers of fluorine, oxygen, and the alpha-particle; M_0, M_α, and M_p are the masses, in grams, of the oxygen atom, alpha-particle and proton; E_d and E_p are the disintegration energy and proton energy, in ergs; $E_p^* = E_p(M_F/(M_F + M_P))$.

Numerically,

$$S=(+8.92/V^{\frac{1}{2}}+28.3/(8.2+V)^{\frac{1}{2}}),\ V \text{ in MEV}.$$

There is thus only one arbitrary constant, k. This constant was calculated to make the curve pass through the point 1.63 MEV and 70. As may be seen from the table, the fit is excellent except for one point that is obviously in error experimentally. It is remarkable that a formula of this character should fit so well, and the fit affords good evidence for the correctness of the underlying assumptions.

TABLE I. *Comparison of theoretical and experimental values for voltage transmutation function of fluorine.*

Voltage (MEV)	Transmutations per 10^9 protons N (Obs.)	N (Calc.)
0.675	0.20	0.34
0.975	2.7	3.7
1.10	4.3	7.7
1.20	13.5	12.8
1.33	23	22.9
1.63	70	70.
0.625		0.20
0.50		0.04
0.25		9×10^{-5}

An error of 50 EKV in determining the energy at the lowest point would make the observed value coincide with the calculated. We were not able to observe scattered protons at less energy than 900 EKV so the energy may well be in error by as much as 50 EKV.

The "threshold" for transmutation seems to be no better defined for fluorine than for lithium

* See Gamow, *Atomic Nuclei and Radioactivity*, p. 97, formula 42.

or boron. Extrapolating the theoretical curve to 500 EKV shows that one would need a current of about 10^{-7} amperes of protons of that energy to detect 10 alpha-particles per minute in a solid angle of $4\pi/60$ (our solid angle), or 50 microamperes at 250 EKV.

From the transmutation function may be calculated a nuclear cross section effective for transmutation. At 1.5 MEV there are about 50 transmutations for an equivalent centimeter of proton track. From the number of fluorine atoms present in such a layer and the stopping power of CaF_2, one may calculate that the effective radius of the fluorine nucleus, within which a proton must strike to produce a transmutation, is 3.5×10^{-14} cm. This value is quite small compared to the nuclear radius deduced from scattering experiments. In other words, the chance of transmutation in a close collision is small. For lithium the corresponding radius is 2×10^{-14} cm.

The Mass of F^{19}

The mechanism suggested by Cockcroft and Walton[1] for this transmutation is ${}_9F^{19}+{}_1H^1={}_8O^{16}+{}_2He^4+E$. The energy available in this reaction, if the mass of the neutral fluorine atom is assumed to be 19.000, is about 5 MEV. Dividing this energy between alpha-particle and recoiling oxygen nucleus gives the alpha-particle 4 MEV, corresponding to a range of about 4.5 cm. This range is consistent with the measurements of Oliphant and Rutherford.[2] However, Bainbridge[13] has shown from his measurements of the mass of Ne^{22} and from the transmutation of fluorine with alpha-particles, as in the experiments of Chadwick and Constable,[14] that the mass of fluorine should be between 19.0022 and 19.0034. If we use our measured alpha-particle range of 6.7 cm at a proton energy of 1.2 MEV, the mass of the fluorine nucleus may be calculated to be 19.0031. This calculation, of course, assumes that no gamma-rays are emitted in the reaction.

In our work the energy of the incident proton is not negligible in comparison to the energy of transmutation. The complete equation for calculating the latter from the measured energy of the alpha-particle and proton is therefore important. This equation is the following:

$$E=\tfrac{1}{2}MU^2+\tfrac{1}{2}MU^2\cdot M/M'-\tfrac{1}{2}mu^2(1-m/M')$$

where m, M and M' are the masses of the proton, alpha-particle, and recoil nucleus, respectively, and u and U are the velocities of the proton and alpha-particle. This equation takes account of the fact that the recoil nucleus must carry off the momentum of the proton and therefore does not recoil in a direction directly opposite to that taken by the alpha-particle. It is calculated on the assumption that the alpha-particle is observed in a direction at right angles to the path of the proton. For lithium the correction term amounts to one-quarter of the proton energy. At low proton energies or higher recoiling masses this correction is not important.

We acknowledge with sincere thanks the continued financial support of the Research Corporation and the Chemical Foundation and the valuable assistance of Commander T. Lucci.

[13] K. Bainbridge, Phys. Rev. **43**, 424 (1933).

[14] J. Chadwick and J. E. R. Constable, Proc. Roy. Soc. **A135**, 48 (1932).