Transmutations of Sodium by Deutons

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Under deuton bombardment sodium emits protons. neutrons and alpha-particles, and becomes radioactive, The radioactive substance, which was shown by chemical tests to be radio-sodium, decays with a half-life of 15.5 hours, emitting electrons with energies up to 1.2 mv, and 5.5±0.5 mv gamma-rays. One microampere of 1.7 mv deutons bombarding a target of NaCl produces about 4×10^6 radio-sodium atoms per second. The rapid increase in yield with voltage predicted by the Gamow formula has been found, and with the higher current and voltage that soon will be available, the already large yield of radio-sodium just stated should be increased more than 100-fold. Doubtless radio-sodium will find many uses in the physical and biological sciences. The number of emitted protons equals, within experimental uncertainty, the number of radio-sodium atoms created; this supports the view that Na^{24} is the radioactive isotope formed. The protons are distributed in ranges up to 49±2 cm (air at 20°C, 760 mm) with a rather pronounced group in the neighborhood of 17 cm. The number of neutrons, emitted presumably in a reaction in which Mg^{24} is formed, is of the same order of magnitude as the number of protons. The alpha-particles have a range of 6.5±0.3 cm and are emitted in a less frequent reaction in which presumably Ne^{21} is formed. Balance of energy in the reactions leads to the following atomic masses:

Na^{23}, 22.992±0.001; Na^{24}, 24.000±0.003;
Mg^{24}, 23.993±0.003.

INTRODUCTION

IN experiments[1] of the character of a preliminary survey, it was observed that some substances under deuton bombardment emit most of the known nuclear radiations—protons, alpha-particles, electrons, positrons, neutrons, gamma-rays—and in addition, become radioactive. These preliminary experiments and the more detailed work of other laboratories[2] made it clear that nuclear reactions induced by deutons are of considerable complexity, and require extensive and detailed study. The present paper is a report of an experimental investigation of some of the reactions of deutons with sodium.

RADIOACTIVITY

The experiments[3] were begun with an investigation of radioactivity induced in sodium by deuton bombardment. For this purpose a beam of high speed deutons from the larger[4] of our instruments for the acceleration of light ions was brought out through an aluminum window into the air. This arrangement has some convenient advantages for studying induced radioactivity. Throughout the experiments a beam of 2.15 mv deutons impinging on the window was used. The range of the deutons in the air outside was observed, by the blue glow throughout their paths, to be 4.7 cm (air at 20°C, 760 mm). By placing a piece of platinum in the path of the beam and observing the range of the scattered deutons with a linear amplifier, it was found that in addition to the main and quite homogeneous group indicated by the glow, there was a relatively small number of considerably greater range, extending to nearly 9 cm. However, the number of these more energetic deutons was so small that they undoubtedly played no important part in the effects here reported.

The procedure was to place sodium 5 mm from the aluminum window in the deuton beam for a given period of time and then to remove the sodium (or sodium-containing material) and place it near an electroscope where the radiations emitted by the radioactive substance in the sodium could be studied.

Although a target having a freshly prepared surface of sodium metal was exposed to the

[1] G. N. Lewis, M. S. Livingston and E. O. Lawrence, Phys. Rev. **44**, 55 (1933); E. O. Lawrence, M. S. Livingston and G. N. Lewis, Phys. Rev. **44**, 56 (1933); E. O. Lawrence and M. S. Livingston, Phys. Rev. **45**, 220 (1934); M. C. Henderson, M. S. Livingston and E. O. Lawrence, Phys. Rev. **45**, 428 (1934).

[2] M. L. E. Oliphant, P. Harteck and Lord Rutherford, Proc. Roy. Soc. **A144**, 692 (1934); J. D. Cockcroft and E. T. S. Walton, Proc. Roy. Soc. **A144**, 704 (1934); H. R. Crane and C. C. Lauritsen, Phys. Rev. **45**, 497 (1934); M. A. Tuve and L. R. Hafstad, Phys. Rev. **45**, 651 (1934).

[3] A preliminary account of some of the results of the experiments on radio-sodium is contained in a letter to the editor, E. O. Lawrence, Phys. Rev. **46**, 746 (1934).

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

deuton beam in order to provide an additional confirmation that sodium atoms were responsible for the activity observed, in most of the experiments a target of sodium chloride, in the form of a crystal of rocksalt, was used. In the first experiments it was found that exposure of the rocksalt to a microampere of deutons for a half hour produced a very large activity. The irradiated crystal, placed near the thin aluminum window of an ionization chamber of 300 cc volume (air at atmospheric pressure), produced an amount of ionization in the chamber, as indicated by the rate of deflection of the fiber of a Lauritsen type electroscope, of the order of magnitude of 10^8 ions per second. In the first few minutes after exposure to the deuton beam, this activity decreased considerably, but after an hour or so the decrease of activity was very much less rapid, and after several hours the activity decayed exponentially with a half-life of about 15 hours. From previous experience[5] it was apparent that the initial short period activity was due to radio-oxygen driven onto the target from the bombarded nitrogen of the surrounding air. In order to eliminate this spurious effect, observations of the sodium activity were taken always many hours after irradiation of the rocksalt, that is, after such a long period of time that the short period activity was reduced to a negligible amount.

Decay Period

In Fig. 1 is shown the variation with time of the radioactivity beginning 20 hours after a one hour exposure of the rocksalt crystal to a beam of 1 microampere of 1.7 mv deutons. The activity is plotted logarithmically, and from the slope of the straight line a half-life of 15.5 ± 0.5 hours is obtained. The precision of the measurements seemingly would warrant setting a smaller limit of error than 0.5 hour. However, it was found that temperature changes appreciably affected the calibration of the electroscope, and since the observations extended over many hours during which no great precautions were taken to keep the temperature constant, this rather large limit of error has been assigned.

The Disintegration Electrons

From the amount of ionization produced in the ionization chamber, the sensitivity of the electroscope, and the geometry of the arrangement, it was possible to estimate approximately the number of beta-particles emitted from the activated rocksalt through the course of time. Thus in Fig. 1 the ordinates represent a logarithmic plot of the absolute number of disintegration electrons emitted per second throughout the whole solid angle 4π resulting from an exposure of 1 hour to 1 microampere of 1.7 mv deutons. Extrapolating back to the time when the salt was exposed to the deuton beam, one obtains an initial yield of 1.6×10^5 disintegration electrons per second. From this value the saturation activity, that is, the activity produced by exposure to the deuton beam for an infinite time, can be calculated. This turns out to be 3.7×10^6 beta-particles per second, and is the rate at which radioactive atoms are formed in the sodium chloride by 1 microampere of 1.7 mv deutons. In other words, one radioactive atom is formed per 1.7×10^6 deutons.

Measurements of the energy of the beta-rays were made by interposing various thicknesses of aluminum sheets between the activated sodium chloride and the ionization chamber. The activated salt was about 3 cm from the ionization

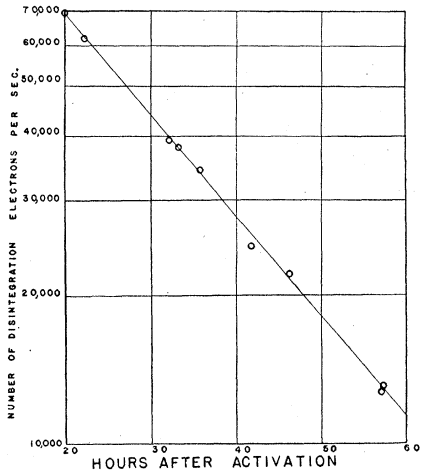

FIG. 1. Radio-sodium decay curve.

[5] M. S. Livingston and E. McMillan (Phys. Rev. **46**, 437 (1934)) have shown that even a platinum target becomes radioactive when exposed to deutons in air in the manner of these experiments, and that the effect is due to radio-oxygen, O^{15}, produced by bombardment of nitrogen.

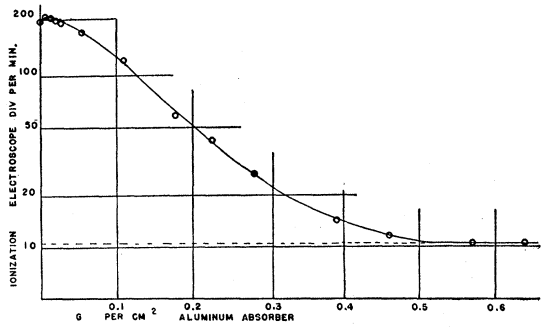

FIG. 2. Beta-ray absorption curve. The dotted line indicates the gamma-ray ionization.

chamber, whose linear dimensions were approximately 10 cm so that the solid angle subtended by the ionization chamber was quite large.

The results are shown in Fig. 2 where the ionization is plotted in essentially arbitrary units, namely rates of deflection of the electroscope fiber in divisions per second, on a logarithmic scale against the thickness of aluminum absorber in grams per sq. cm. The ionization falls off with increase in aluminum absorber to a constant value of about 1/20 of the initial value. Clearly the ionization is produced by both beta and gamma-rays and an indication of the maximum energy of the beta-rays is obtained by noting the thickness of aluminum at which the ionization becomes approximately independent of the amount of aluminum absorber. Thus the estimated maximum range of the beta-particles is about 0.51 gram per sq. cm.

Using Feather's[6] empirical formula for the relation between the maximum energy of beta-particles and their maximum range

$$E=(R+0.091)/0.511,$$

where R is in grams per sq. cm of aluminum, and the energy E is expressed in millions of electron volts, the observed range of the beta-particles indicates that their maximum energy is about 1.2 mv. In making an estimate of the maximum energy of beta-particles in this way, the tendency would of course be to estimate too low a value for the endpoint of the range, and therefore too low a value for the maximum energy. It remains for further and more extensive measurements, probably by magnetic deflections, to determine the endpoint with greater precision. However, the upper limit to the energy of the disintegration electrons is here determined with enough precision to show that the Sargent[7] relation holds. This beta-disintegration fits on the Sargent curve for Ra E and Th C.

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

RADIOACTIVE GAMMA-RAYS

From the relative amount of beta and gamma-ionization, the relative number of beta and gamma-rays can be estimated as follows. Using the same electroscope and ionization chamber, McMillan[8] has studied the annihilation radiation produced in activated carbon. He found that this 0.5 mv radiation gave rise to about 1/40 of the ionization produced by the positive electrons from the activated carbon. In the case of activated carbon (radio-nitrogen) it is to be expected, as indeed has been demonstrated approximately by Lauritsen and Crane,[9] that there should be twice as many gamma-quanta as positive electrons. In the present experiments several measurements of the ratio of gamma to beta-ionization gave values ranging from 1/33 to 1/20. If the radioactive sodium radiation were of the same energy as the annihilation radiation from N^{13}, one would conclude then that in this case more than two quanta accompany each beta disintegration. However, as will be seen below, gamma-rays from activated sodium have much higher energy (5.5 mv), and give more ionization per quantum passing through the ionization chamber. In view of this, the observations indicate that, within the uncertainty of the experiments, equal numbers of beta and gamma-rays are given off from radio-sodium. In other words, one gamma-ray accompanies each radioactive disintegration.

In order to determine the energy of the gamma-ray, absorption measurements were made in aluminum, copper and lead. For this purpose there was used another ionization chamber built especially for such absorption measurements and employed by McMillan[8] in his experiments on gamma-rays from various substances. The essential geometry of the arrangement is shown in

[7] B. W. Sargent, Proc. Roy. Soc. **A139**, 659 (1933).
[8] E. McMillan, Phys. Rev. **46**, 868 (1934).
[9] H. R. Crane and C. C. Lauritsen, Phys. Rev. **45**, 430 (1934).

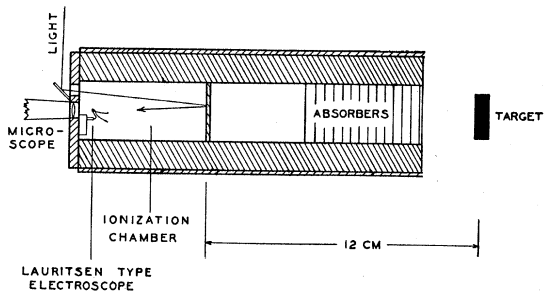

FIG. 3. Arrangement of ionization chamber and absorbers. The electroscope consists of a sputtered quartz fiber supported at one end on a light wire frame. A short cross-hair attached to the free end of the fiber is observed against a scale in the microscope eyepiece.

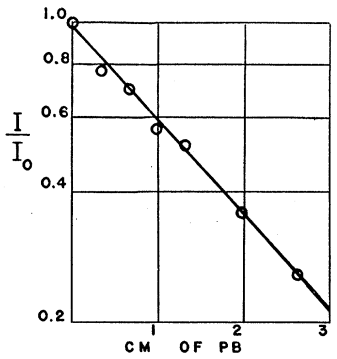

FIG. 4. Absorption in Pb of gamma-rays from radio-sodium.

Fig. 3. The cylindrical lead-lined ionization chamber was 2.5 cm in diameter and 5.7 cm long. The absorbers consisted of disks of various thicknesses, placed relative to the ionization chamber and to the source of gamma-rays, as shown in the diagram. The distance from the ionization chamber to the activated sodium was 12 cm.

The results of the absorption measurements in lead are shown in Fig. 4 where the relative ionization produced by the gamma-rays is plotted logarithmically as a function of the thickness of the lead absorber. Within experimental uncertainty the absorption is exponential, indicating that the radiation is monochromatic, with an apparent absorption coefficient of 0.51 cm^{-1}.

According to the theory of pair production (materialization of radiation) of Oppenheimer and Plesset[10] and of Bethe and Heitler,[11] which has been verified experimentally by McMillan[8] and by Lauritsen and Crane,[12] the absorption coefficient of the gamma-rays in lead does not uniquely determine the quantum energy, and it is necessary to make observations of absorption in lighter elements where pair production is not so important. Accordingly the apparent absorption coefficients of the radiation in copper and aluminum were observed, and the values 0.275 cm^{-1} and 0.066 cm^{-1}, respectively, were obtained.

Forward scattering of the radiation in the absorber influences the observed absorption coefficients, and it is a difficult matter to make proper corrections. Dr. McMillan calibrated the ionization chamber arrangement with highly filtered gamma-radiation from radium and found that the observed absorption coefficient was 6 percent less than the accepted value. For elements of low atomic number where pair production is not important, this correction is greater for more energetic gamma-radiation because the scattered radiation becomes greater in the forward direction. This increase in the scattering correction can be calculated from the geometry of source, absorber and ionization chamber on the basis of the Klein-Nishina formula. For elements of high atomic number and for radiation of five million volts or more, pair production absorption accounts for more than half of the total absorption, so that the correction for scattering in the case of heavy elements is correspondingly reduced.

With these considerations in mind, corrections to the observed absorption coefficients in aluminum, copper and lead have been made of 6, 5 and 3 percent, respectively. These corrected values,[13] given in terms of absorption per electron, are plotted in Fig. 5 against the atomic number of the absorber. A consideration of various sources of uncertainty in the measurements leads to the conclusion that these values are probably right to within 10 percent. The two straight lines represent respectively the

[10] J. R. Oppenheimer and M. S. Plesset, Phys. Rev. **44**, 53 (1933).

[11] Bethe and Heitler, Proc. Roy. Soc. **A146**, 83 (1934).

[12] H. R. Crane and C. C. Lauritsen, Int. Conf. Phys., London, Oct., 1934.

[13] According to a theoretical calculation by Hall (H. Hall, Phys. Rev. **45**, 216 (1934); H. Hall and W. Rarita, Phys. Rev. **46**, 143 (1934)) photoelectric absorption of 5 mv radiation is negligible in Al and Cu but in Pb it amounts to 4.3 percent of the total. A correction of this amount has accordingly been made in the plot of Fig. 5.

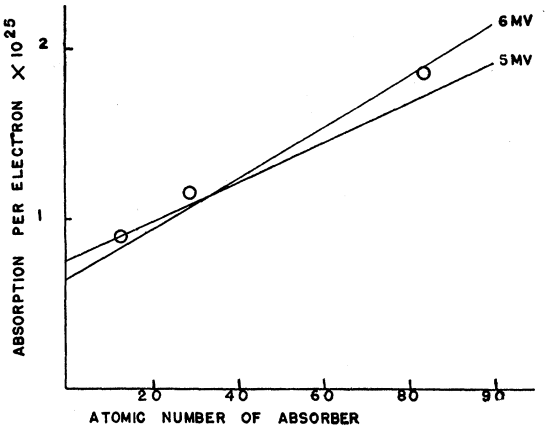

FIG. 5. The variation with atomic number of the absorption per electron of the radio-sodium gamma-rays.

variation with atomic number for 5 and 6 mv radiation of the total absorption, i.e., Klein-Nishina plus pair production, according to calculations supplied the writer some time ago by Professor Oppenheimer.[14] The observations show clearly that the absorption per electron in lead is more than twice as great as that in aluminum, and that therefore the radiation is of high energy. But on the basis of the absorption measurements alone, it is not possible to estimate with precision the quantum energy of the radiation. The extrapolated value for the absorption at zero atomic number indicates that the radiation is in the neighborhood of 5 mv, whereas the rate of increase of absorption with atomic number suggests radiation of about 6 mv. The best estimate from the absorption data has therefore been taken as 5.5 ± 0.5 mv.

Presumably the gamma-ray is emitted following the beta-disintegration, that is to say, from $_{12}Mg^{24}$ (see below) in an excited state. It is noteworthy that gamma-rays of about this energy have also been observed[15] from C^{12} and O^{16}. These three nuclei are of the same type in the sense that they can be regarded as made up of alpha-particles alone, and in view of this, it perhaps is not surprising that they should exhibit similar excitation levels.

[14] The writer is much indebted to Professor Oppenheimer for this information.

[15] H. Becker and W. Bothe, Zeits. f. Physik **76**, 421 (1932). E. McMillan, reference 8.

CHEMICAL TESTS

The atomic number of the radioactive element, which manifestly could not be far removed from that of sodium was determined by the following chemical tests. An activated sample of sodium metal together with some aluminum and magnesium was dissolved in concentrated sulfuric acid and then evaporated to dryness. The activity remained in the residue, indicating that the radioactive substance was certainly not fluorine or neon. The precipitate was next dissolved in water with the addition of a little ammonia. Phosphate was then added, which precipitated the magnesium and the aluminum. The activity was found to remain in the solution, showing that almost certainly the active substance was sodium. Fermi[16] observed radioactivity of 15 hours half-life induced in aluminum by neutrons, and he also came to the conclusion from chemical tests that the radioactive substance is a sodium isotope. In view of the identity of the observed half-lives, there can be little doubt that in both cases the same radioactive isotope was formed.

This chemical proof that radio-sodium is formed by the reaction of deutons with ordinary sodium suggests at once the following nuclear reaction:

$$_{11}Na^{23}+{}_1D^2\rightarrow{}_{11}Na^{24}+{}_1H^1. \qquad (1)$$

THE PROTONS

The protons[17] which, according to the above reaction, accompany the production of radio-sodium, have been observed and studied in some detail as follows.

A carefully cleaned crystal of rocksalt was mounted in vacuum and bombarded with a few hundredths of a microampere of 2.15 mv deutons. Protons in a solid angle of $4\pi/200$ at right angles

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

[17] Evidence of these protons was already at hand in the early experiments of Lawrence, Livingston and Lewis (reference 1). They found that several groups of protons, including one of 35 cm range, were emitted from a target of sodium phosphate bombarded by 1.3 mv deutons. At that time it was not established whether the groups were due to sodium or phosphorus or both (oxygen was excluded by experiments with other targets), but it seemed likely that at least one of the groups was due to sodium. In the present experiments it is evident that the 35 cm group was in fact due to this element.

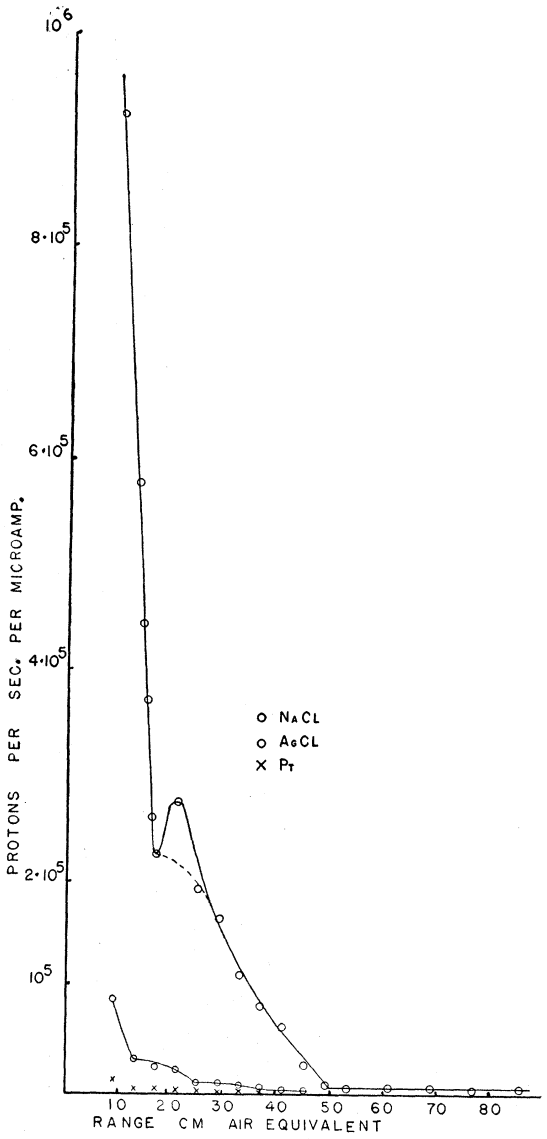

FIG. 6. Range distribution of protons emitted from Na under deuton bombardment.

to the deuton beam, passed through a mica window and through various absorbers into an ionization chamber 1 cm deep, where the ionization pulses produced by the protons were counted by a linear amplifier-thyratron-scale-of-8 arrangement. The bias voltage on the thyratrons was such that only those protons were counted that were near the end of their range when in the ionization chamber. For the most part, aluminum foils of 0.001 inch thickness were used for the absorption measurements. Calibration with polonium alpha-particles showed that they had a stopping power of 4 cm of air at 20°C and 760 mm.[18] For variations of the absorber less than 4 cm, air itself was used. The number of proton counts from the rocksalt target as a function of the thickness of the absorber is shown in Fig. 6. Although in the measurements it was necessary to use deuton currents of less than 1/10 of a microampere in order to record individual proton counts, the ordinates of Fig. 6 give the number of protons emitted per second in every direction per microampere of 2.15 mv deutons, which was readily calculated from the observational data.

Although considerable care was exercised with regard to the cleanliness of the targets, there existed a likelihood that an appreciable part of the observed proton emission from NaCl was due to contamination, particularly as the rocksalt was exposed to air before being placed in position in the vacuum chamber. Oxygen and nitrogen especially were suspects. The probable presence of a thin film of carbonaceous material from the vapor of the oil diffusion pumps was also recognized. However, a large amount of carbon contamination could be excluded on the basis of the range of the protons. From the observations that under deuton bombardment carbon emits protons having a range of 14 ± 1 cm at 0.5 mv[19] and 18 ± 1 cm at 1.2 mv,[20] it is to be expected that at 2.15 mv their range would be 24 cm. The peak in the proton range curve at 21 cm is close to this value and might conceivably be due to a slight amount of carbon contaminant. This possibility is noted in the figure by the dotted line.

In order to examine the constituents of air (particularly oxygen and nitrogen) as possible contaminants, observations were made of the proton emission from the sodium chloride target when bombarded with 1.2 mv deutons in air; following which the sodium chloride target was removed from the deuton beam and the proton emission from the air alone was observed. From

[18] All range measurements in this paper refer to air at 20°C, 760 mm.
[19] J. D. Cockcroft and E. T. S. Walton, Proc. Roy. Soc. **A144**, 704 (1934).
[20] E. O. Lawrence, M. S. Livingston and G. N. Lewis, Phys. Rev. **44**, 56 (1933).

the air two proton groups were observed, of ranges 86 and 26 cm, and in addition a copious emission of alpha-particles having ranges of 7.6 and 13.4 cm, respectively. By replacing the air atmosphere with oxygen, it was established that the great majority of these protons and alpha-particles were not due to oxygen and therefore, in all likelihood, were due to nitrogen.[21]

But quite apart from the question of the origin of the several groups of particles from air, a proton group having a range of 38 ± 2 cm was found to be present only when the sodium chloride target was bombarded. When account was taken of the lower deuton energy in the air experiments, this 38 cm group corresponded well with the 49 ± 2 cm group (see Fig. 6) observed in the vacuum experiments. Therefore the evidence strongly indicated that the 49 cm group was due to sodium chloride. The 17 ± 1 cm group observed in vacuum could only tentatively be ascribed to sodium chloride, inasmuch as these particles would have had a range in the air experiments of only 12 cm, and for this reason it would have been difficult to distinguish them in the presence of the large alpha-particle emission.

In order to distinguish between chlorine and sodium, a target of silver chloride was bombarded in vacuum, with 2.15 mv deutons, with the results also shown in Fig. 6. From this target three definite proton groups of ranges approximately 13, 23 and 38 cm, respectively, were observed. These are probably to be ascribed to chlorine, but require further investigation. However, since the protons emitted from the sodium chloride target were very much greater in number, they are certainly not to be ascribed to chlorine. Finally, in order to expose a possible unsuspected contaminant under the conditions of the experiment, the NaCl target was replaced by a platinum target in vacuum, and proton counts were made with the same deuton current. The very small number of counts shown in Fig. 6 was accounted for as due either to scattered deutons or to the general neutron background. The evidence therefore strongly indicates that the observed emission of protons from the sodium chloride target is to be ascribed to sodium.

It should be emphasized that the data of Fig. 6 represent a range distribution rather than an integrated range curve, inasmuch as only those protons were counted that were very near the end of their range. In order to estimate the total yield of disintegration protons, it was necessary therefore to determine how near to the end of its range a proton had to be in order to be counted. This was accomplished in the following manner. Protons emitted at right angles to the deuton beam in air were collimated by slits 3 mm wide and 3 cm apart, so that protons that passed through the two slits came from an air target less than 4 mm thick. In this manner a fairly homogeneous group of long range protons was obtained to test the performance of the amplifier. On the assumption that the spread in energy of the nitrogen protons was equal to that of the bombarding deutons over the thickness of the air target, it was calculated that the spread in ranges of these nitrogen disintegration protons was about 2 cm, while it was found that the linear amplifier counted the majority over a range interval of 4 cm. Hence it was concluded that in order for a proton to be counted, it had to pass through the linear amplifier within 2 cm of the end of its range. Thus the ordinates of the curves shown in Fig. 6 represent the number per second of disintegration protons in a range interval of 2 cm emitted in all directions per microampere.

If the protons are produced in the nuclear reaction in which radio-sodium is formed, it follows that the number of protons emitted per second should equal the number of radioactive atoms formed per second. This may be checked by numerically integrating the proton counts of Fig. 6, assuming that the linear amplifier counts effectively over a range interval of 2 cm. The rate of emission of protons with ranges greater than 9 cm is estimated in that way to be 5×10^6 protons per second per microampere. Scattered deutons prevented determination of the proton range distribution below 9 cm and extrapolation of the curve is necessarily uncertain. However, a reasonable extrapolation gives a total proton yield of 15×10^6 protons per sec. This is to be compared to the estimated rate of production of

[21] We expect to submit for publication in the not distant future an account of these experiments on the disintegration of nitrogen by deutons.

radioactive sodium atoms (i.e., the saturation beta-activity), which is 3.7×10^6 per second. The protons were produced by 2.15 mv deutons, while the estimate for the saturation radioactivity is for 1.7 mv deutons, and from the transmutation function, it is expected that the yield in the former case should be about five times that in the latter. Thus within experimental uncertainty the observations indicate that the yield of protons equals the yield of radio-sodium atoms, in support of the assumed reaction.

Finally, mention should be made of two aspects of the observed distribution of the emitted protons. First, the fact that at least two proton groups of ranges about 17 cm (3.4 mv) and 49 cm (6.2 mv), respectively, seem to be emitted from sodium, suggests that a gamma-ray is often emitted in the nuclear reaction with an energy corresponding to the difference in energy of the two groups which is, taking account of the energy of recoil of Mg, 3.9 mv. Second, the range distribution of the groups seems to be greater than is to be accounted for by the thick target effect, and it is not impossible that this wide energy distribution is related to the energy distribution of the disintegration electrons. These are matters that remain to be examined more closely later on.[22]

Differential Transmutation Function

In order to study the amount of the induced activity as a function of the energy of the bombarding deutons, a target consisting of several thin mica foils (each having a stopping power of about 1 cm air equivalent) was exposed to the deuton beam. Mica contains, among other things, sodium, and the variation of the sodium activity with energy of the bombarding deutons was determined by observing the amount of the

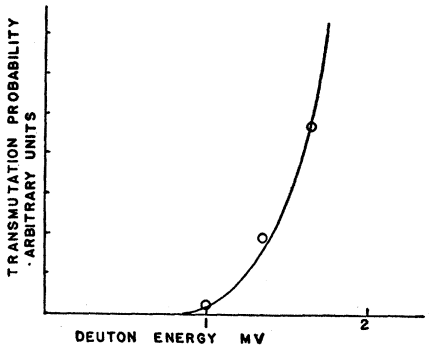

FIG. 7. The variation with deuton energy of the sodium radioactivity produced in a thin target.

15 hour activity produced in the several mica foils. The foil nearest the aluminum window exhibited the largest activity because the deutons had the greatest average velocity in passing through it. Thus the amount of the activity was observed to decrease rapidly with succeeding foils, and the observations yielded what might be termed a differential transmutation function, or thin target transmutation function. This manner of measuring the transmutation function has the advantage that it is unnecessary to take account of the magnitude of the current of bombarding deutons or of the time of exposure, as the integrated exposures of all the thin targets are automatically the same.

The activity decreased so rapidly with energy of the deutons that an appreciable effect was observed in only the first three mica foils. These results are plotted in Fig. 7. The ordinates represent the activity, necessarily on an arbitrary scale, and are proportional to the probability of activation of the sodium by the deutons for various energies in millions of volt-electrons as given by the abscissas.

The curve is a plot of the Gamow function

$$N=K(1/v)e^{-4\pi^2Ze^2/hv}.$$

N is the relative probability that a deuton of velocity v will penetrate the potential barrier of a nucleus of atomic number Z. The arbitrary constant K has been adjusted to the best fit with the experimental data.

Thus, as can be seen from this plot, the variation with deuton energy of the induced activity is accounted for on the assumption that

[22] Dr. McMillan and I have been studying the reactions of deutons with aluminum, and among other things find that aluminum[28] is formed with the emission of protons in a quite analogous manner to the sodium reaction here discussed. The range distribution of the protons from aluminum is quite similar to that of sodium. However, in the case of aluminum, we have been able to obtain a high resolving power by using thin targets and narrow collimating slits, and find that the apparent continuous range distribution in fact consists of a super-position of many proton groups. It therefore appears not unlikely that the wide distribution in ranges of the protons from sodium is due to many proton groups.

the all-important factor is the ability of the deuton to penetrate the barrier of the sodium nucleus. We have found this to be true also in the case of aluminum. Indeed, in every case so far examined carefully in our laboratory, the Gamow theory accounts for the variation of the probability of a nuclear reaction with the energy of the bombarding particle.

Utility of Radio-Sodium

Radio-sodium will doubtless find many uses because it has a conveniently long life, and can be produced in large amounts. In the present experiments with 1 microampere of 2.15 mv deutons, an amount of radio-sodium was produced which gave an activity of more than 10^7 beta-particles per second. The yield of radio-sodium increases very rapidly with the voltage and with currents and voltages that will soon be available, it will be possible to produce quantities of radio-sodium more than a hundred times greater than here reported.

The very energetic gamma-radiation, which is presumably monochromatic makes radio-sodium an ideal gamma-ray source for certain experimental studies. Already a more precise experimental investigation of the absorption of the radiation in various elements has been undertaken in our laboratory, the results of which should have important bearing on the theory of the Klein-Nishina and pair production absorption. Additional experiments using the Wilson chamber are planned, from which it is hoped it will be possible to gain more detailed information about the processes of both beta and gamma-absorption.

Radio-sodium is particularly suited for many biological uses because if injected into biological material it would leave after a few days activity an innocuous non-radioactive residue.

Neutrons

Since the end products of the reaction involving radio-sodium are Mg^{24}, H^1 and an electron, it seemed likely that an alternative reaction occurs in which sodium is converted directly into Mg^{24} with the emission of a neutron, *viz.:*

$$_{11}Na^{23}+{}_1D^2\rightarrow{}_{12}Mg^{24}+{}_0n^1. \qquad (2)$$

Neutron observations were made difficult by the rather high neutron background produced by deutons striking various parts of the apparatus other than the target under investigation. However, by making observations with the deutons electrostatically deflected alternately on and off the target, the background could be allowed for in qualitative fashion. In addition, a piece of Be mounted on a ground joint could be moved into the deuton beam directly in front of the target so that the neutron emission from the target could be compared with the emission from Be.

For the neutron observations a block of paraffin was placed directly in front of the ionization chamber and the recoil protons ejected by the neutrons were counted. The ionization chamber in these experiments subtended a solid angle at the target of about $4\pi/100$. Using a current of deutons of 0.04 microampere, the proton counts per minute were Be, 750; NaCl, 96; and the background, 48. Similar observations with a AgCl target using 0.1 microampere gave the following counts per minute: Be, 1840; AgCl, 112; and the background, 128. Neutron emission was thus definitely measurable above the background in the case of NaCl, but not in the case of AgCl. A third experiment with a platinum target gave, as expected, no effect above the background.

Thus, the experiments showed that under deuton bombardment Na emits neutrons. A rough estimate of the neutron yield may be made on the assumption that 1 recoil proton was recorded on the average for every 1000 neutrons passing through the ionization chamber. On this assumption, which is certainly correct in order of magnitude, it is calculated from the experimental data that the neutron yield from a NaCl target bombarded by 2.15 mv deutons is about 3 per 10^7 deutons. This nuclear reaction therefore has a probability of the same order of magnitude as the one in which radio-sodium is formed.

The Alpha-Particles

Observations of the ionization pulses with a cathode-ray oscillograph showed that in addition to the protons and neutrons, a considerable number of alpha-particles were emitted from

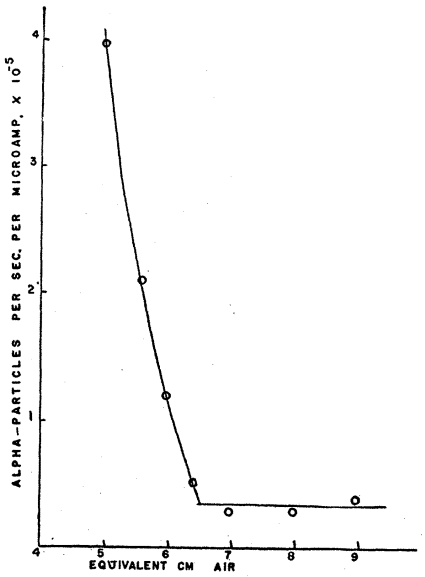

FIG. 8. Range distribution of alpha-particles from Na under deuton bombardment.

sodium chloride. From silver chloride, however, the alpha-particle emission was of a smaller order of magnitude, and it therefore was concluded that the alpha-particle emission in the former case was ascribable to sodium. In making range measurements, the alpha-particles were distinguished from the protons by increasing threefold the bias voltage on the thyratron counter. Throughout these measurements care was taken to keep the bombarding currents within a magnitude such that there was no appreciable piling up of proton pulses, which would give spurious counts. The results, reduced to the total yield per microampere of 2.15 mv deutons, are plotted in Fig. 8. The particles have a range of 6.5 ± 0.3 cm. The counts recorded in the case of a greater thickness of absorber were due to the neutrons both from the sodium and the general background radiation.

The reaction here concerned presumably is:

$$_{11}Na^{23}+{}_1D^2\rightarrow{}_{10}Ne^{21}+{}_2He^4.$$

The total yield of alpha-particles per second from sodium chloride bombarded by 2.15 mv deutons was calculated from the data to be 1 per 10^7 deutons. Thus the probability of this third reaction is smaller than that of either of the first two. According to the Gamow theory, this result is quite reasonable, since the probability that a nuclear reaction of this type will occur depends not only on the probability that a bombarding particle will get in, but also on the probability that a nuclear particle will get out. Because the nuclear barrier for the alpha-particle is greater than that for the proton, it follows that one would expect a greater probability for the proton reaction.

Energy Balance and Atomic Masses

By balancing energy in the reactions, the masses of Na^{23}, Na^{24} and Mg^{24} may be determined from the experimental data and known mass values as follows. If no gamma-radiation is given off in the alpha-particle reaction, we have

$$_{11}Na^{23}+{}_1D^2+(\text{k.e. of }{}_1D^2)={}_{10}Ne^{21}+{}_2He^4+(\text{k.e. of }{}_2He^4)+(\text{k.e. of }{}_{10}Ne^{21}).$$

The kinetic energy of the deutons was 2.15 mv ($=0.0023$ mass unit) and the alpha-particles had a range of 6.5 cm corresponding to 7.3 mv ($=0.0078$ mass unit), and it follows from conservation of momentum that the recoil kinetic energy of the Ne^{21} was 0.0017 mass unit. Taking Aston's value for He, 4.0022, and Bainbridge's value for H^2, 2.0136, and Bainbridge's provisional value[23] for Ne^{21}, 20.996 ± 0.001, we have accordingly

$$Na^{23}=20.996\pm0.001+4.0022+0.0078+0.0017-2.0136-0.0023$$
$$=22.9918^{22}\pm0.001.$$

This value for the mass of Na^{23} is not in satisfactory agreement with that derived from its chemical atomic weight,[24] i.e., 22.999 ± 0.003. In view of the possibility that gamma-rays are given off in the above reaction, the chemical value is more trustworthy.

Likewise, if it is assumed that no gamma-radiation is given off in the reaction in which radio-sodium is formed, we have

[23] The writer is much indebted to Dr. Bainbridge for this information.

[24] I am indebted to Professor Birge for calling my attention to the recent determinations by C. R. Johnson (J. Phys. Chem. **37**, 923 (1933) and Baxter and Hale (J. Am. Chem. Soc. **56**, 615 (1934), which indicate that sodium has a chemical atomic weight of 22.994 ± 0.002, which when reduced to the $O^{16}=16$ scale becomes 22.999 ± 0.003.

$_{11}Na^{23}+{}_1D^2+$(k.e. of D^2)
$=Na^{24}+H^1+$(k.e. of H^1)$+$(k.e. of Na^{24}).

Using the chemical value for Na^{23}, and the observed maximum proton energy, 6.2 mv (=0.0067 mass unit), we obtain

$$_{11}Na^{24}=22.999\pm0.003+2.0136+0.0023-1.0078-0.0067-0.0006$$
$$=24.000\pm0.003.$$

Finally if the difference in mass between Na^{24} and Mg^{24} is represented by the sum of the maximum energy of the beta-rays (1.2 mv) and the energy of the gamma-rays, 5.5 ± 0.5 mv, we have

$$Mg^{24}=24.000\pm0.003-0.0013-0.0059$$
$$=23.993\pm0.003.$$

It should be noted that the assumed relation between the maximum energy of the beta-particles and the mass change in the beta-disintegration has not heretofore been established by experiment or justified by theory. The fact that the value for the mass of Mg^{24} obtained in this way fits satisfactorily on Aston's mass defect curve therefore may be interpreted as experimental evidence favoring the validity of this fundamental assumption.

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Comparison of the Spectra of CaH and CaD

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The spectrum of calcium deuteride has been photographed at high dispersion with a Ca arc in an atmosphere of heavy hydrogen as a source. Quantum analyses of the B and C systems are presented, and the rotational energy constants and spin doubling of the two states of the B bands are compared with those for CaH. The ratio $B_e{}^i/B_e$ for the ground state is 0.51337 whereas the ratio of the reduced masses is 0.51276. This represents a decrease of 0.059 percent for the equilibrium internuclear distance in CaD. The spin doubling in the $B\,{}^2\Sigma$ state of CaD does not show the irregular variation with K found in CaH, and is of magnitude almost exactly in the ratio ρ^2 with that of the Λ-doubling in the $A\,{}^2\Pi$ state of CaH. Multiple large perturbations occurring in the $C\,{}^2\Sigma$ state of CaD are described.

INTRODUCTION

IN the several band systems of the CaH molecule are to be found striking examples of the phenomena of l-uncoupling, Λ-doubling, predissociation and perturbations.[1] The magnitudes of all of these effects depend upon the rotational constants B_v and the relative spacing of the rotational energy levels in the interacting states. Substitution of deuterium for hydrogen in this molecule therefore produces, because of the relatively large change in the reduced mass, interesting variations in these phenomena. In addition, the study of the hydrogen isotope effect in these bands provides an example for the prediction by Kronig[2] that in certain cases the potential energy function for the vibration of the non-rotating molecule is not identical for the two isotopes, the equilibrium distance being slightly smaller for the heavier isotope.

EXPERIMENTAL PROCEDURE

The light source was a d.c. arc carrying 2.5 amperes between a water-cooled copper anode and a hollow copper cathode filled with metallic

[1] For references cf. W. Jevons, *Report on Band-Spectra of Diatomic Molecules.*

[2] R. de L. Kronig, Physica **1**, 617 (1934).