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The Energy released in the Disintegration of Nitrogen by Thermal Neutrons*

By

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ABSTRACT

The energy released in the ${}_7N^{14}(n,p){}_6C^{14}$ reaction produced by thermal neutrons has been determined by the parallel plates ionization chamber with linear amplifier filled with atmospheric air dried by $CaCl_2$. For the energy calibration, alpha-particles from ThC′ were used as standard, and the correction for the systematic error introduced by the calibration was made by the aid of the theoretical analysis of the pulse shape in the linear amplifier. By assuming the proportionality between the amount of the total ionization in air and the energy of the particle, we have obtained the Q-value of the reaction as 0.609 ± 0.005 Mev. Taking the end point energy of the beta-spectrum of ${}_6C^{14}$ as 0.156 ± 0.001 Mev, the neutron-hydrogen mass difference has been determined as 0.765 ± 0.005 Mev.

1. Introduction

The reaction of ${}_7N^{14}$ produced by thermal neutrons

$$ {}_7N^{14} + {}_0n^1 = {}_6C^{14} + {}_1H^1 + Q $$

was investigated in the earlier experiments by Chadwick and Goldhaber(1), Bonner and Brubaker (2, 3), and Burcham and Goldhaber (4). The final nucleus ${}_6C^{14}$ emits a negative electron and returns to the initial nucleus ${}_7N^{14}$ again ;

$$ {}_6C^{14} = {}_7N^{14} + E_\beta . $$

Here E_β is the extrapolated end point energy of the beta-ray spectrum.**

Bonner and Brubaker (3) first pointed out that the neutron-hydrogen mass difference can be obtained from the above disintegration cycle ;

$$ n - H = Q + E_\beta $$

* This paper was read at the meeting of Nuclear Physics Division, Physical Society of Japan, April 2, 1950.

** As pointed out by Kofoed-Hansen(5), the extrapolated Fermi end point involves the neutrino rest mass if any.

In 1947, Stephens (6) made a synthetic discussion on the experimental studies relating to the neutron-hydrogen mass difference. The most reliable result available in 1947 was the value $n - \mathrm{H} = 0.755 \pm 0.016$ Mev obtained from the photo-disintegration of deuterium (7). The results obtained from various kind of disintegration cycles, though they were somewhat inaccurate, gave, on the average, $n - \mathrm{H} = 0.757$ Mev (6) in agreement with the photo-disintegration data. And the consistency of the results seemed to be satisfactory to that time.

Notwithstanding, however, Bell and Elliott (8) made in 1948 an experiment determining the binding energy of deuterium by measuring the gamma-ray energy from ${}_1\mathrm{H}^1(n, \gamma){}_1\mathrm{D}^2$, and obtained the value $n - \mathrm{H} = 0.804 \pm 0.009$ Mev which was about 50 Kev higher than the previously accepted value. The discrepancy was too large to be explained by the experimental errors concerned.

Under these circumstances, a keen interest has arisen for the more accurate redetermination of the binding energy of deuterium and the neutron-hydrogen mass difference. In view of the roughness of the existing data on the disintegration cycles, it has been especially desirable to investigate carefully these cycles.

As for the nitrogen cycle above mentioned, the end point energy of the beta-ray from ${}_6\mathrm{C}^{14}$ has been accurately and consistently determined by several experiments (9), while the Q-values given by various investigators for the ${}_7\mathrm{N}^{14}(n, p){}_6\mathrm{C}^{14}$ reaction are fairly inconsistent. The results obtained by the ionization measurements are : 0.57 ± 0.04 Mev by Huber, Huber and Scherrer (10), 0.63 ± 0.01 Mev by Huber and Stebler (11), and 0.71 Mev by Barshall and Battat (12). Cloud chamber observations of this reaction have given, however, well consistent values of proton range, i. e., 1.00 cm (13). If we assume Bethe's range-energy relation (14) to be correct in this region, we have a Q-value of 0.60 Mev.

In the present experiment, we have attempted to determine the Q-value of the ${}_7\mathrm{N}^{14}(n, p){}_6\mathrm{C}^{14}$ reaction by the method of measuring the amount of total ionization as accurately as possible.

2. Experimental procedure and apparatus

The fundamental difficulties encountered in the accurate determination of the nuclear reaction energy by measuring the total ionization produced by the particles in an ionization chamber with linear amplifier are related to the following problems.

The first and more essential is the relation between the amount of the total ionization and the energy of the particle. For the purpose

of the present study, the generally accepted law of proportionality between these quantities may be considered to be somewhat daring postulation, since the law has been derived from experiments of less accuracy. Up to the present time, the exact characters of this relationship in various sorts of gases and for different natures of particles have not been studied with sufficient accuracy. Thus, we cannot but assume it for the present.

The second is concerned with the technical problems to insure the proportionality between the total ionization produced in the ionization chamber and the output pulse height of the linear amplifier. In general, the output pulse height due to a fixed total ionization produced by a particle of fixed energy (regardless of the statistical fluctuation of the ionization process) is not necessarily constant but varies corresponding to the location of the disintegrated nucleus and the direction of the emitted particle in the ionization chamber. The degree of this inhomogeneity depends upon the relative magnitudes of the time constant of the amplifier and the collecting time of ions, the latter of which is defined by the mobility of ions and the chamber voltage.

In order to guarantee the proportionality between the ionization and the pulse height, the time constant of the amplifier should be adequately large in comparison with the collecting time of ions. Since the practical limitation of the collecting time is set by the high voltage source and the chamber dimension, the amplifier should be operated with large time constant to acquire good linearity. On the other hand, this makes it difficult to reduce the low frequency noise of the amplifier such as microphonics and mechanical vibrations.

Indeed, this difficulty is overcome in principle by adopting the technique of the electron collection first introduced by Ortner(**15**), since the mobility of electrons in pure gases is high and the collecting time can be made sufficiently small. But, owing to the serious location effect of the tracks in the chamber, simple ionization chamber of parallel plates type cannot be used adequately. If we use, then, chambers with screen grid or chambers of cylindrical or spherical shape, it is not so easy to insure the strict proportionality between the ionization and the pulse height, though approximately it is attained. In the case of ionization chamber with screen grid, high shielding effect of the screen grid is desirable for the elimination of the above-mentioned location effect, but at the same time it promotes the capture of electrons by the grid, and it is fairly difficult to find an adequate compromise.* For the cylindrical

* The detailed discussion on this subject was reported at the meeting of Nuclear Physics Division, Physical Society of Japan, April 3, 1950.

or spherical chambers, the homogeneity attained is of essentially approximate character and the estimation of the degree of inhomogeneity by theoretical analysis is utterly troublesome unless the range of the particle is negligibly small in comparison with the chamber dimension. Thus it seems that the electron collection technique has not necessarily the decisive superiority for our present purpose.

As a consequence, we have adopted an ionization chamber of parallel plates type operated under conditions of non electron collection, and chosen the chamber voltage as high as practicable and the amplifier time constant small enough to suppress the low frequency noise. The deviation from the proportionality between the ionization and the pulse height was analysed theoretically, and the correction was made to the observed result as described later.

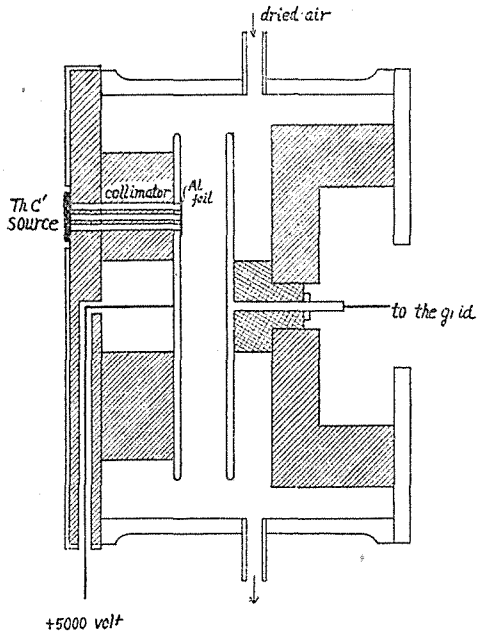

Fig. 1. The ionization chamber used.

Fig. 1 shows the ionization chamber used. It consists of a parallel plates condenser made of two copper disks of 2 mm thickness and 11.3 cm diameter (about 100 cm^2 area). The depth of the chamber was designed to be about 1.5 cm after the knowledge of the proton range 1.00 cm (**13**) of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction in N. T. P. air, and the actual depth proved to be 1.473 ± 0.006 cm by the accurate measurement. It was a good compromise between the disadvantage of the so-called wall effect and the demand for short collecting time, since the voltage required for a certain collecting time is proportional to the square of the chamber depth. Actually + 5000 volt was applied to the high voltage electrode from a well-stabilized source, since this voltage was found to be the highest voltage practicable under the ordinary room humidity. The high voltage electrode has several small holes of 2 mm diameter to admit alpha-particles for the energy calibration. These holes are covered with thin aluminium foil to prevent the distortion of the field.

Now, instead of using pure nitrogen, the air of atmospheric pressure and dried by $CaCl_2$ was filled in the chamber, because the mobility of the negative ions is much greater than that of positive ions and its value is very variable when ordinary tank nitrogen is used.* The reason for the selection of air dried by $CaCl_2$ is simply that in the preliminary experiment the positive and the negative ions in such dried air have been found to have the same mobility, $k_+ = k_- = 2.2\ \mathrm{cm^2/sec/volt}$,** under the present conditions of dryness and age of ions (age $< 2.5 \times 10^{-4}$ sec), and that the theoretical calculation becomes, then, simplified by this knowledge. The existence of oxygen nuclei does not interfere the experiment, because the Q-values for the (n, p) and (n, α) reactions of oxygen are expected to be negative.

The amplifier used is a conventional resistance-capacity coupled type with four stages. This is a modification of the circuit developed by K. Kimura and Y. Uemura (**16**) in our laboratory. Great care was taken for the selection of the time constant which was small enough to suppress the noise and the gamma-ray disturbance from the Ra + Be neutron source but not so small as to distort the linearity seriously. After several trials and theoretical calculations, we have chosen the value of 1.68×10^{-3} sec for the clipping time constant (between the first and second stages) and much greater value of 1.22×10^{-2} sec for the later stages. For the determination of the values of the time constants, capacities and resistances were directly measured by the bridge method, and the plate resistances were obtained from the static characteristics of the tubes.

When usual cares for preventing the low frequency noise such as the use of rubber cushions and the fuse metal as lead wires and etc. were taken, the above combination was found to be satisfactory for the reduction of noise disturbances and yield a good resolution. The ratio of the clipping time constant to the ion collecting time (1.95×10^{-4} sec) is about 8.6. This ratio is not large enough to insure the strict proportionality between the ionization and the pulse height. The distortion, however, is not serious and the correction requested is only about 1.7 per cent as discussed later.

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* Experiments are still in progress.

** The tabulated values of mobilities for air, $k_+ = 1.6$ and $k_- = 2.2$, were found in the preliminary experiment to be valid for the ions whose age is longer than 1×10^{-3} sec. These results on the ion mobility for air together with other results were reported at the meeting of Nuclear Physics Division, Physical Society of Japan, April 3, 1950, and will be soon published in this Memoirs.

The input tube used was UZ-6001* and selected carefully among many tubes of the same kind. This was operated with floating grid for the purpose of making the time constant of the input system properly greater than about one second. Under these circumstances, the distortion of the pulse shape in the input stage was negligibly small, and the possible cause of distortion was confined to the interstage coupling circuits.

For the adjustment of the gain and the linearity of the amplifier, a thyratron pulse generator similar to that used by Holloway and Livingston (**17**) was used. This pulse generator proved to have satisfactory precision if the voltage source was well stabilized.

The energy calibration of the pulse height was carried out by the comparison with the pulses of alpha-particles from ThC′. The ThC′ source deposited on a small copper disk was attached to the holes bored in the outer wall of the chamber, and alpha-particles were admitted into the chamber through collimators of 36 mm length and 2 mm diameter. The distance between the source and the inner surface of the high voltage electrode was 36.021 ± 0.055 mm, and the air equivalent of the aluminium foil was 2.82 ± 0.02 mm.

Pulses were recorded by photographing the screen of an oscilloscope of 14 cm diameter, and the pulse heights were directly measured on the films. Fig. 2 shows the linearity of the over-all system including amplifier, oscilloscope and camera measured with the above-mentioned pulse generator.

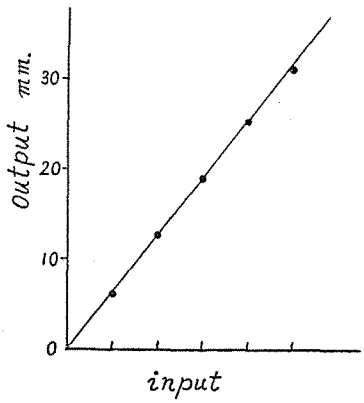

Fig. 2. The linearity of the over-all system.

The neutron source used was a 50 mg Ra + Be mixture. The geometrical arrangement of the chamber and the source is shown in Fig. 3. For the absorption of the gamma-ray from the source, a lead block of about 12 cm was used. About 10 cm of paraffin were also placed between the chamber and the source to avoid fast neutrons which would make recoil atoms in the chamber and interfere the statistics.

* Non-microphonic and low noise level pentode manufactured by Tokyo Shibaura Electric Co. Ltd., and with the same characteristics as UZ-77.

Under these geometrical conditions, apparently uniform pulses attributed to the ${}_7N^{14}(n, p){}_6C^{14}$ reaction produced by thermal neutrons were obtained at a rate of about fifteen per minutes.

In such a low counting rate, the so-called back-kick effect on the pulse height distribution is negligibly small. The time of each of the photographic exposures was from one to two minutes.

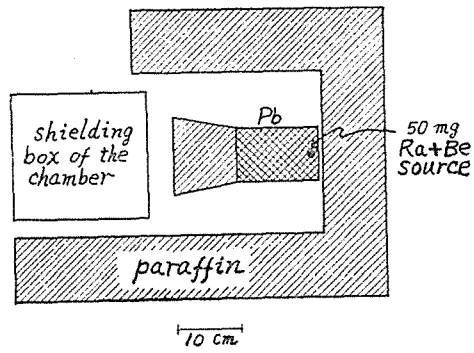

Fig. 3. The geometrical arrangement of the chamber and the neutron source.

3. Results

About one thousand pulses due to the ${}_7N^{14}(n, p){}_6C^{14}$ reaction were recorded. Fig. 4 shows the distribution in size of these pulses measured on the photographic films. The dotted curve is the distribution of the pulses of the pulse generator attached to check the gain of the amplifier. The symmetrical broadening of this curve is due to the non-systematic noise of the amplifier and to that of the gamma-ray disturbance. It is obviously approximated by Gaussian distribution. The standard deviation of this distribution, i.e., the r.m.s. value of the noise, is about 0.9 mm, and it corresponds to about 27 Kev of the input ionization energy. The width of the distribution of the ${}_7N^{14}(n, p){}_6C^{14}$ pulses is the superposition of the main noise broadening and the small amount of inhomogeneity due to the location effect of ${}_7N^{14}$ nuclei in the chamber.

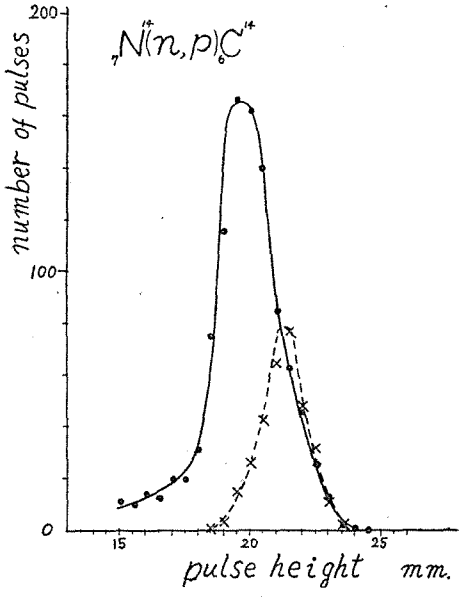

Fig. 4. The distribution of the pulses due to the ${}_7N^{14}(n, p){}_6C^{14}$ reaction. The dotted curve is the distribution of the pulses of the pulse generator showing the noise broadening.

Fig. 5 shows the distribution of the ThC′ alpha-pulses for the calibration. In this observation, the gain of the amplifier was diminished by a factor of about two in such a way that the alpha-pulses have almost the same magnitude as that of the pulses due to the ${}_7N^{14}(n, p){}_6C^{14}$ reaction. It was aimed at minimizing the error introduced by the possible distortion of the linearity of the whole recording system, though Fig. 2 showed a good linearity.

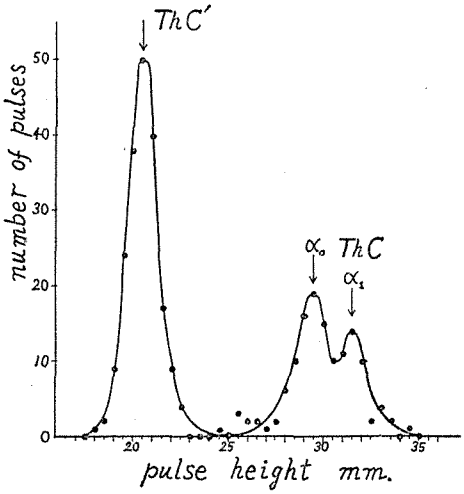

Fig. 5. The distribution of the Th(C+C′) alpha-pulses for the energy calibration.

The exact rate of the change of the gain was obtained from the measurement of the ratio of the resistances of the potential divider in the pulse generator, and the corresponding signal pulse heights in both cases. As shown in the figure, the ThC alpha-groups, α_0 and α_1, are clearly discriminated showing good resolution. For the energy calibration, the main peak of ThC′ was used.

The room temperature and the atmospheric pressure were constant throughout the observation, namely 22.3° C and 755.1 mm Hg respectively.

The energy lost by ThC′ alpha-particle in the chamber was determined by the range-energy curve of Holloway and Livingston (**17**). The situations are shown in Fig. 6 ; R_0 is the mean range of ThC′alpha-particle, b the distance between the source and the inner surface of the high voltage electrode (corrected for temperature and pressure to fit the range-energy curve in N. T. P.), a the air equivalent of the aluminium foil, and d the depth of the chamber (also corrected for temperature and pressure). The energy E_0 lost by the alpha-particle in the chamber is obtained as the difference of the energies E_1 and E_2 corresponding to the points A and B as shown in the figure. The fact that the distance b was so chosen that the alpha-particle is not stopped in the chamber but hit against the collecting electrode with adequate energy, is favourable to minimize the error introduced in the estimation of E_0.

First, if we take the value 8.570 ± 0.007 cm (17) for the mean range of ThC′ and the values already mentioned for b, a, d, and temperature and pressure, the values E_1 and E_2 are obtained as 6.124 and 4.866 Mev respectively from the range-energy curve. Thus only the most accurate region of the range-energy curve (5～9 Mev) is concerned with the estimation of E_0. Second, since we take the difference of the energies corresponding to two points on the range-energy curve, the uncertainties involved in the values R_0, a, and b take their effect on the final error through the difference of dE/dR at A and B. This also minimize the error as compared with the case when the alpha-particle is stopped in the chamber and the final error is directly connected with dE/dR at A. After all, the main sources of error accompanying estimation of E_0 are the uncertainty involved in the value of the chamber depth d and the errors introduced in the procedures of reading the graph. The error arising from the imperfection of the collimation of alpha-particles is negligibly small. After these careful considerations and procedures, the energy lost by ThC′ alpha-particle in the chamber was determined as

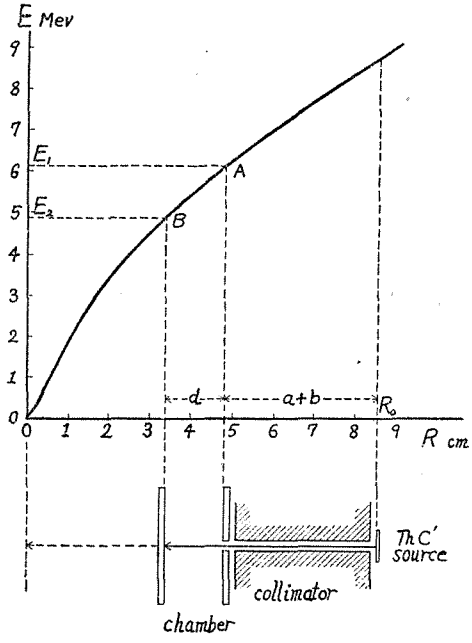

Fig. 6. The illustration of the estimation of the energy lost by ThC′ alpha-particle in the chamber.

$$E_0 = 1.258 \pm 0.006 \text{ Mev},$$

where the amount of the error attached was estimated by taking account of all sources of possible errors above mentioned.

In order to determine the Q-value of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction from the distribution curves shown in Figs. 4 and 5, we must consider here about the correction for the distortion of the proportionality between the ionization and the pulse height as mentioned in the preceding section.

Now, let C be the capacity of the input system (including the chamber, lead wire, and the grid of the input tube) and q be the charge of the ions produced in the chamber, then the output pulse height h is generally expressed by

$$h = Afq/C,$$

where, A denotes the linear amplification factor* of the amplifier, and f the reduction factor of the pulse height due to the distortion of the pulse shape in the interstage coupling circuits. The value of the factor f depends upon the relative magnitudes of the ion collecting time and the time constant of the amplifier, and also the ionization density along the path of the particle. Thus, for certain values of the amplifier time constant and the applied voltage, the value of f varies with the location and the direction of the tracks of the disintegrated particle in the chamber.

Therefore, for the observation of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction in which the disintegrated nuclei are distributed throughout the chamber, the variation of the f-value will exhibit a certain distribution around the average reduction factor $\bar{f}_N$. On the other hand, the alpha-particles used for the energy calibration are admitted into the chamber with a fixed geometry, so that in this case the factor f will take a definite value f_T, which is in general different from $\bar{f}_N$. As a consequence, in the determination of the Q-value by the comparison of the pulse heights obtained from Figs. 4 and 5, the correction must be made for the discrepancy of $\bar{f}_N$ and f_T**.

We had made a theoretical analysis*** of the pulse shape and the reduction factor f in the ionization chamber of parallel plates type, and this analysis was applied to the evaluation of the numerical values of $\bar{f}_N$ and f_T for the present experiment. In this calculation, the proton range of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction was taken as 1.00 cm (**13**) in N.T.P. air and the charge density along the path of the proton was approximated by a polynomial of the second degree to fit Bethe's range-energy relation. Ionizations shared between the proton and the recoil

* Under the present experimental conditions, vacuum tube itself can be regarded as a linear element of the circuit.

** It is noticeable that the correction required for the distortion of the proportionality between the ionization and the pulse height is not a definite factor characteristic to the apparatus, but the discrepancy of the reduction factors in both the real observation and the calibration.

*** The details of this analysis and its experimental verification were reported at the meeting of Nuclear Physics Division, Physical Society of Japan, April 3, 1950, and will be published in this Memoirs.

nucleus ${}_6C^{14}$ were assumed to be inversely proportional to the masses, and the range of ${}_6C^{14}$ was neglected in comparison with the chamber dimension and treated as a point charge. For ThC′ alpha-particle, the charge density along the path was also properly approximated to fit the range-energy curve of Holloway and Livingston. The effect of temperature and pressure and the distortions in the later coupling stages were also taken into account.

In Fig. 7 the calculated output pulse height under the present experimental conditions is plotted against the location of the disintegrated ${}_7N^{14}$ and the direction of the emitted proton. The abscissa represents

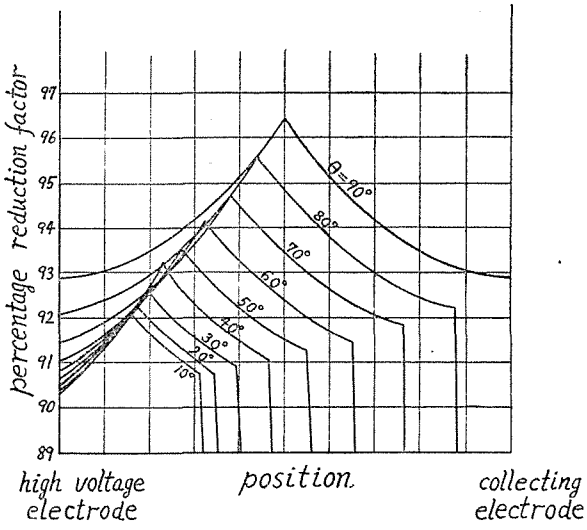

Fig. 7. The percentage reduction factor plotted against the position of the ${}_7N^{14}$ nucleus and the direction of the emitted proton in the chamber.

the position of the disintegrated nucleus in the chamber and θ denotes the angle between the direction of proton and the normal to the electrode plate. The ordinate shows the percentage reduction factor. Each curve shows the variation of f corresponding to fixed θ and various positions. In the case when $180^\circ > \theta > 90^\circ$, curves are turned over symmetrically to the vertical line drawn at the middle point of the abscissa in consequence of the fact that $k_+ = k_-$. The rapid drops in the right-hand sides of the curves represent the so-called wall-effect, i. e., the effect of the protons which are stopped by the electrode with some residual ranges.

By assuming that the occurrences of the reaction are uniformly distributed in the chamber and the directions of the emitted protons are spherically symmetrical, the distribution of the f-value in the observation of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction can be obtained from the curves in Fig. 7.

Fig. 8 shows the resulting distribution of the f-value obtained by this means* (curves in Fig. 7 having been calculated for the interval of five degrees). The mean value $\bar{f}_N$ of the theoretical distribution is 92.6 per cent. In the figure, the calculated value of f_T for the calibration with ThC′ alpha-particle is also shown by a thick vertical line at 91.0 per cent.

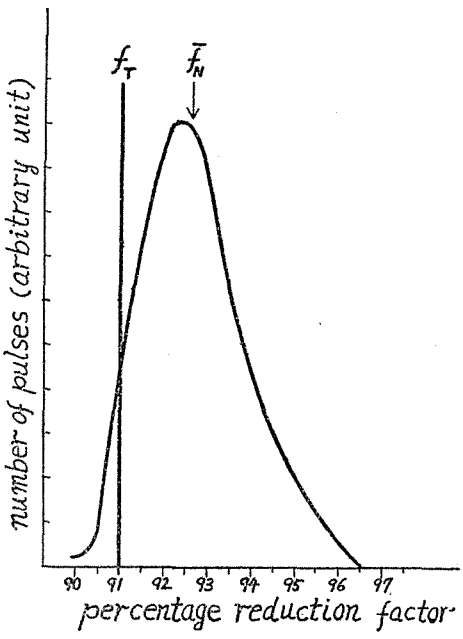

Fig. 8. The calculated distribution of f-value for the observation of the ${}_7N^{14}(n, p){}_6C^{14}$ reaction and the f-value for the calibration with ThC′ alpha-particle.

As already mentioned, the observed distribution is overlapped by the broadening due to noise and gamma-ray disturbance which composes the main part of the actual broadening**. However, if we take the mean value in the determination of the pulse height from the observed distribution, the effect of the noise broadening on the result will be eliminated automatically owing to its essentially symmetric character. Thus the above obtained factor 91.0/92.6 can be properly used for the correction***, even if the noise broadening composes the main part of the actual broadening.

* In this distribution, the edge-effect of the ionization chamber was also taken into account by assuming that the effective volume of the chamber is a clearly defined cylinder whose radius is equal to that of the electrode. Other assumptions were also made on the field distribution near the edge of the effective volume, but they yielded almost the same distributions.

** The half-maximum width for the theoretical distribution of the ${}_7N^{14}(n, p){}_6C^{14}$ pulses is only about 2.8 % of the mean value $\bar{f}_N$. In the observed distributions, this ratio amounts to 11.5 % for ${}_7N^{14}(n, p){}_6C^{14}$ and 8.8 % for ThC′.

*** There is no anxiety for the columnar recombination in the present experiment. According to Moulin's experiment (18) on the columnar recombination, the practical saturation is attained in atmospheric air at about 500 volt/cm. The field of 3400 volt/cm in the present experiment seems to insure the perfect saturation for the columnar recombination.

Now, if we assume the strict proportionality between the total ionization and the energy for both alpha-particle and proton, the Q-value for the ${}_7N^{14}(n, p){}_6C^{14}$ reaction is given by the following expression:

$$Q = E_0 \frac{R_2 S_T N f_T}{R_1 T S_N \bar{f}_N}.$$

In this expression, E_0 is the energy lost by ThC′ alpha-particle in the chamber; R_1 and R_2 the resistances in the potential divider of the pulse generator; T and N the observed pulse heights for ThC′ and the ${}_7N^{14}(n, p){}_6C^{14}$ reaction respectively; S_T and S_N the pulse heights from the pulse generator measured to check the gain of the amplifier in both cases; and f_T and $\bar{f}_N$ are the reduction factors already discussed. The numerical values of these factors have been determined as follows:

$$E_0 = 1.258 \pm 0.006 \text{ Mev},$$
$$R_1 = 4094.4 \pm 1.0 \text{ ohm}, \quad R_2 = 2081.4 \pm 0.7 \text{ ohm},$$
$$N = 19.98 \pm 0.04 \text{ mm}, \quad S_N = 21.26 \pm 0.05 \text{ mm},$$
$$T = 20.48 \pm 0.06 \text{ mm}, \quad S_T = 21.11 \pm 0.07 \text{ mm},$$
$$f_T = 91.0\ \%, \quad \frac{f_T}{\bar{f}_N} = 0.983 \pm 0.002.$$
$$\bar{f}_N = 92.6\ \%,$$

The error accompanying the estimation of f-value is mainly due to the uncertainty of the ion collecting time, and the contribution of the uncertainty of the amplifier time constant is negligibly small in the present experiment. If we take the following values as the factors defining the collecting time; the chamber depth $d = 1.473 \pm 0.006$ cm, the ion mobility $k_+ = k_- = 2.2 \pm 0.1$, and the applied voltage $V = 5000 \pm 100$ volt, the uncertainty existing in the estimation of f-value becomes about 0.4 per cent. However, the effects of above uncertainties of the values d, k, and V on each value of f_T and $\bar{f}_N$ are not independent, but there is a direct correlation between them. Therefore, the final error for the ratio $f_T/\bar{f}_N$ is considered to be much smaller than 0.4 per cent. The above indicated error for this ratio is the upper limit that may be conceivable.

Putting these values in the above expression, we obtain the Q-value for the ${}_7N^{14}(n, p){}_6C^{14}$ reaction:

$$Q = 0.609 \pm 0.005 \text{ Mev}.$$

The probable error attached here does not include any possible error involved in the assumption on the proportionality between the ionization and the energy.

4. Discussion

By taking the extrapolated end point of ${}_6C^{14}$ beta-ray as 0.156 ± 0.001 Mev (19), the neutron-hydrogen mass difference is estimated to be

$$n - H = 0.765 \pm 0.005 \text{ Mev}$$

from the present result. This value is in agreement with the value 0.755 ± 0.016 Mev obtained from the photo-disintegration of deuterium within the limits of experimental errors, and definitely smaller than the value 0.804 ± 0.009 Mev obtained by Bell and Elliott (8).

Recently, Stephens and his collaborators (20) made a simultaneous measurement of the Q-values for ${}_7N^{14}(n,p){}_6C^{14}$ and ${}_2He^3(n,p){}_1T^3$ reactions with ionization chamber filled with mixture of argon, nitrogen and helium, and determined the average $n - H$ value as 0.785 ± 0.006 Mev. In their experiment, Q-value obtained for ${}_7N^{14}(n,p){}_6C^{14}$ was 0.630 ± 0.006 Mev. This is about 20 Kev higher than the result of the present experiment. The threshold measurement of the inverse reaction ${}_6C^{14}(p,n){}_7N^{14}$ was also made recently by Shoupp, Jennings and Sun (21). They obtained the Q-value as 0.620 ± 0.009 Mev which gave rise to the $n - H$ value of 0.776 ± 0.009 Mev. Within the limit of experimental error, this value is consistent with the result of the present experiment as well as with that of Stephens's experiments.

For the ionization measurement including the present experiment and that of Stephens, the fundamental assumption is the proportionality between the total ionization and the energy for both alpha-particle and proton in question. Up to the present time, however, this assumption has not acquired sufficient experimental support. Jesse, Forstat and Sadauskis (22) have recently examined the ionization-energy relation in argon for alpha-particle. Their result maintains the strict proportionality in argon for alpha-particle of energy 1.5 to 9 Mev. In their report, they also discussed about the proportionality in air by converting Stetter's result (23) of the ionization-range relation for alpha-particle into the ionization-energy relation, but this latter argument does not seem so reliable as their experimental result in argon. Moreover, no reliable experiments are available for proton at all. After all, it is difficult to find a proper explanation for the above discrepancy.

Meanwhile, in recent times fairly accurate determinations of the neutron-hydrogen mass difference have been made from other aspects of this subject. Tollestrup, Jenkins, Fowler and Lauritsen (24) obtained 0.789 ± 0.006 Mev from the measurement of Q-values for D-D reactions. Taschek, Argo, Hemmendinger and Jarvis (25) obtained 0.782 ± 0.002

Mev from the threshold measurement of the ${}_1T^3(p, n){}_2He^3$ reaction. These results together with that of Stephens are about 20 Kev smaller than the value of Bell and Elliott, and also about 20 Kev higher than the present result.

Though these values are in good agreement with each other, threshold measurements involve in general uncertainties arising from the voltage calibration and the nature of the yield curve near the threshold point. Therefore, it is not reasonable to discuss here merits and demerits of these results and the value of the present result and the values (7) determined from the photo-disintegration of deuterium observed by K. Kimura, Myers and van Atta, and Wiedenbeck and Marhoefer. Further examination of the ionization-energy relation and the more accurate redetermination of the binding energy of deuterium are desirable for clearing up the confusion.

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REFERENCES

1. J. Chadwick and M. Goldhaber, Proc. Camb. Phil. Soc., **31** (1935), 612.
2. T. W. Bonner and W. M. Brubaker, Phys. Rev., **49** (1936), 223.
3. T. W. Bonner and W. M. Brubaker, Phys. Rev., **49** (1936), 778.
4. W. E. Burcham and M. Goldhaber, Proc. Camb. Phil. Soc., **32** (1936), 632.
5. O. Kofoed-Hansen, Phys. Rev., **71** (1947), 451.
6. W. E. Stephens, Rev. Mod. Phys., **19** (1947), 19.
7. G. Stetter and W. Jentschke, Zeits. f. Phys., **110** (1938), 214.
   K. Kimura, Mem. Coll. Sci. Kyoto Univ., **A 22** (1939), 237.
   F. E. Myers and L. C. van Atta, Phys. Rev., **61** (1942), 19.
   M. L. Wiedenbeck and C. J. Marhoefer, Phys. Rev., **67** (1945), 54.
8. R. E. Bell and L. G. Elliott, Phys. Rev., **74** (1948), 1552.
9. P. W. Levy, Phys. Rev., **72** (1947), 248.
   A. K. Solomon, R. G. Gould and C. B. Anfinsen, Phys. Rev., **72** (1947), 1097.
   C. S. Cook, L. M. Langer and H. C. Price, Phys. Rev., **74** (1948), 548.
10. O. Huber, P. Huber and P. Scherrer, Helv. Phys. Acta, **XIII** (1940) 209.
11. P. Huber and A. Stebler. Phys. Rev., **73** (1948), 85.
12. H. H. Barshall and M. E. Battat, Phys. Rev., **70** (1946), 245.
13. J. K. Bøggild, Kgl. Danske Vid. Sels. Mat.-Fys. Medd., **23** (1945), 22.
    D. J. Hughes and C. Eggler, Phys. Rev., **73** (1948), 809.
    I. C. Cornog, W. Franzen and W. E. Stephens, Phys. Rev., **74** (1948), 1.

14. M. S. Livingston and H. A. Bethe, Rev. Mod. Phys., **9** (1937), 268.
15. G. Ortner and G. Stetter, Phys. Zeits., **35** (1934), 563.
G. Ortner and G. Protiwinsky, Win. Ber., **148** (1939), 349.
16. K. Kimura and Y. Uemura, Mem. Coll. Sci. Kyoto Univ,, **A 23** (1940), 1.
17. M. G. Holloway and M. S. Livingston, Phys. Rev., **54** (1938), 18.
18. L. B. Loeb, *Fundamental Processes of Electrical Discharge in Gases.* (John Wiley & Sons, 1939), 141.
19. Cook, Langer and Price, *loc. cit.*, ref. 9.
20. W. Franzen, J. Halpern and W. E. Stephens, Phys. Rev., **77** (1950), 641.
21. W. E. Shoupp, B. Jennings and K. H. Sun, Phys. Rev., **75** (1949), 1.
22. W. P. Jesse, H. Forstat and J. Sadauskis, Phys. Rev., **77** (1950), 782.
23. G. Stetter, Zeits. f. Phys., **120** (1943), 639.
24. A. V. Tollestrup, F. A. Jenkins, W. A. Fowler and C. C. Lauritsen, Phys. Rev., **75** (1949), 1947.
25. R. F. Taschek, H. V. Argo, A. Hemmendinger and G. A. Jarvis, Phys. Rev., **76** (1949), 325.

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