Further Observations on the Production of N^{13}

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(Received January 5, 1938)

The reactions $C^{12}+H^1\rightarrow N^{13}+h\nu$ and $C^{12}+H^2\rightarrow N^{13}+n^1$ have been studied and the absolute yield of positrons measured for various voltages. The proton-capture reaction shows a single resonance of 30 kv half-width at 450 kv. This resonance has been compared with the similar lithium resonance to fit it into the voltage scale of the lithium and fluorine resonances. Comparison of the positron yields of the two reactions permits a calculation of the yield of N^{13} in the proton reaction from the yield of neutrons in the deuteron reaction as measured by Amaldi, Hafstad, and Tuve. The yield thus determined is approximately 7.5 times higher than the previously accepted value based on positron measurements, and corresponds to a total reaction cross section of about 8.4×10^{-24} volt cm^2. However, the observed number of positrons from the deuteron reaction, obtained by comparison with uranium, is 2.5 times less than the number of neutrons from the same reaction. This discrepancy could be explained by K electron capture by the N^{13} nucleus, but the errors involved are too uncertain to say definitely that this process does occur. An unsuccessful attempt was made to observe scattering of protons by carbon at the resonance voltage.

INTRODUCTION

SEVERAL problems of considerable interest and theoretical importance are connected with the production of N^{13} by proton and deuteron bombardment of carbon ($C^{12}+H^1\rightarrow N^{13}+h\nu$, $C^{12}+H^2\rightarrow N^{13}+n^1$). The early work of Hafstad and Tuve[1] indicated a possible doublet-structure in the resonance capture of protons, but the scatter of their points was such that its existence was doubtful. Their work on carbon was done prior to the installation of the high resistance voltmeter. It seemed desirable to repeat the measurement of the carbon resonance for proton capture to fit it into the voltage scale established by the lithium and fluorine resonances.[2] An anomalous scattering of protons at the resonance energy is expected in elements having a broad resonance for radiative capture, and carbon seemed a favorable element for the detection of this anomaly because of its availability in a gaseous form, its low atomic number, and the large resonance-width established by our measurements as reported below.

The production of N^{13} by deuteron bombardment of carbon brings up another interesting problem. In this reaction each N^{13} atom produced is accompanied by a neutron, but the absolute yield of neutrons measured by Amaldi, Hafstad, and Tuve[3] is approximately seven times larger than the yield of positrons reported by Hafstad and Tuve. If these figures were sufficiently accurate, they would constitute evidence for K electron capture with a high probability by the N^{13} nucleus, but, while the neutron yield has an estimated error of 20 percent, the early measurements of the positron yield may be in error by a rather large unknown factor due to the former uncertainties regarding voltage measurements. We have therefore tried to improve the measurement of the yield of positrons to such a degree that if any large discrepancy exists between the yield of neutrons and positrons it should be attributed definitely to K electron capture.

PROCEDURE

Carbon targets, consisting of one-inch disks of Acheson graphite, were activated by two-minute exposure to the ion beam in vacuum. The activity was measured by observing the initial and final times for a standard deflection of a string electrometer connected to an ionization chamber of the Amaldi-Fermi type[4] containing oxygen at a pressure of three atmospheres and having a window of 0.1 mm aluminum. Immediately after each measurement the sensitivity of the electrometer was checked by observing the time required for the same deflection to be produced by a calibrated uranium beta-ray source. A

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[1] Hafstad and Tuve, Phys. Rev. **48**, 306–315 (1935).
[2] Hafstad, Heydenburg and Tuve, Phys. Rev. **50**, 504–514 (1936).
[3] Amaldi, Hafstad and Tuve, Phys. Rev. **51**, 896–912 (1937).
[4] Amaldi and Fermi, Phys. Rev. **50**, 899–928 (1936).

nomogram calculated from the period of radionitrogen (taken to be 620 seconds) was used to compute what fraction of the total activity had been observed. Each observation then gave the activity in terms of the intensity of the uranium source and independent of the sensitivity of the electrometer. Drift corrections were negligible except for the lowest voltage points.

The accuracy of the nomogram, that is, the correctness of the period selected, was determined by making two such observations with each of a group of ten targets. The activities calculated from the two sets of data agreed to within one percent although the initial times of the observations were roughly one and six minutes after activation. As the initial time for each subsequent observation was very close to one minute and the sensitivity of electrometer was adjusted to give approximately constant deflection time, in spite of large variations of target activity, the error arising in the use of the nomogram was negligible.

For the determination of the absolute yield, the electrometer was calibrated in terms of uranium beta-particles per division by the standardized uranium source, which had been previously used by Amaldi, Hafstad, and Tuve for measurement of neutron intensities.[3] The beta-particles from this source had to penetrate 0.2 mm of aluminum before reaching the ionization chamber, so the ionization was produced solely by the beta-particles from UX_2. In converting the calibration to positrons per division no correction was made for any difference in ionizing power as the beta-ray spectrum of UX_2 is similar to that of N^{13}.[5–7] Neither was any correction made for reflection of the beta-rays, as the target and the uranium standard had approximately the same atomic number and mass per square centimeter, the standard consisting of a small mass of UNO_3 embedded in a much larger mass of paraffin and wrapped in 0.1 mm of aluminum. It is reasonable to believe that the neglect of these factors does not introduce an error as great as 50 percent.

Two other sources of error occur, one in the measurement of the ion current and one in the loss of the radioactive gas from the target. From previous experience the error in ion-current measurement (due to secondary electron effects and contamination of the magnetically analyzed ion beam) is known to be less than five percent. The loss of radioactive gas while the target was in the air was shown to be negligible by the appearance of the decay curve. The loss of gas while the target was in the vacuum was estimated by evacuating a target of known activity. After four minutes of evacuation the activity was 10 percent less than would be expected from the normal decay. In the ordinary procedure the targets remained in the vacuum for two minutes during activation and approximately 20 seconds after activation. Consequently a reasonable estimate of the loss would be five percent. Variations in these two errors are believed to be mainly responsible for the scatter of the points shown in Fig. 1. A correction was made for the decay of the target during bombardment.

Voltage Determination

The high resistance voltmeter was originally calibrated in 1935.[2] Last April the rubber tubing in which the resistors were enclosed was replaced by varnished cambric tubing, about one dozen of the individual resistors which differed from 10 megohms by as much as 10 percent being renewed at the same time. It was also uncertain whether high humidity (early part of October) might have an appreciable effect on the calibration. It therefore seemed necessary to make a recalibration of the voltmeter before we could establish accurately the voltage of the proton-capture resonance in carbon. This was done by observing the lithium gamma-ray resonance at 440 kv. Since this voltage for the lithium resonance was obtained immediately after the original calibration of the voltmeter and has been checked in other laboratories,[8, 9] it is thought to be essentially correct.[10] A repetition of this gamma-ray

[5] Kurie, Richardson and Paxton, Phys. Rev. **49**, 368–381 (1936).
[6] Froler, Delsasso and Lauritsen, Phys. Rev. **49**, 561–574 (1936).
[7] Sargent, Proc. Roy. Soc. **A139**, 659–673 (1933).
[8] Bothe and Gentner, Zeits. f. Physik **104**, 685–693 (1937).
[9] Herb, Kerst and McKibben, Phys. Rev. **51**, 691–698 (1937).
[10] Measurements of the Coulomb scattering of protons by nitrogen in this laboratory provide an independent absolute scale of voltage which agrees with the lithium calibration within less than two percent, the limit of accuracy originally ascribed to the absolute values on the lithium-fluorine resonance scale.

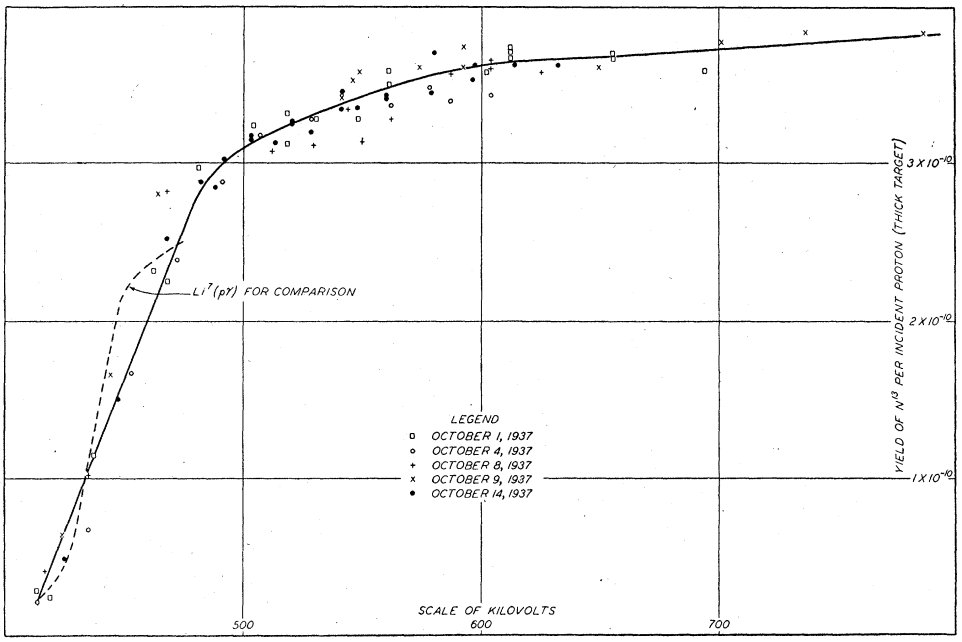

FIG. 1. Production of N^{13} by protons. $C^{12}+H^1\rightarrow N^{13}+h\nu$.

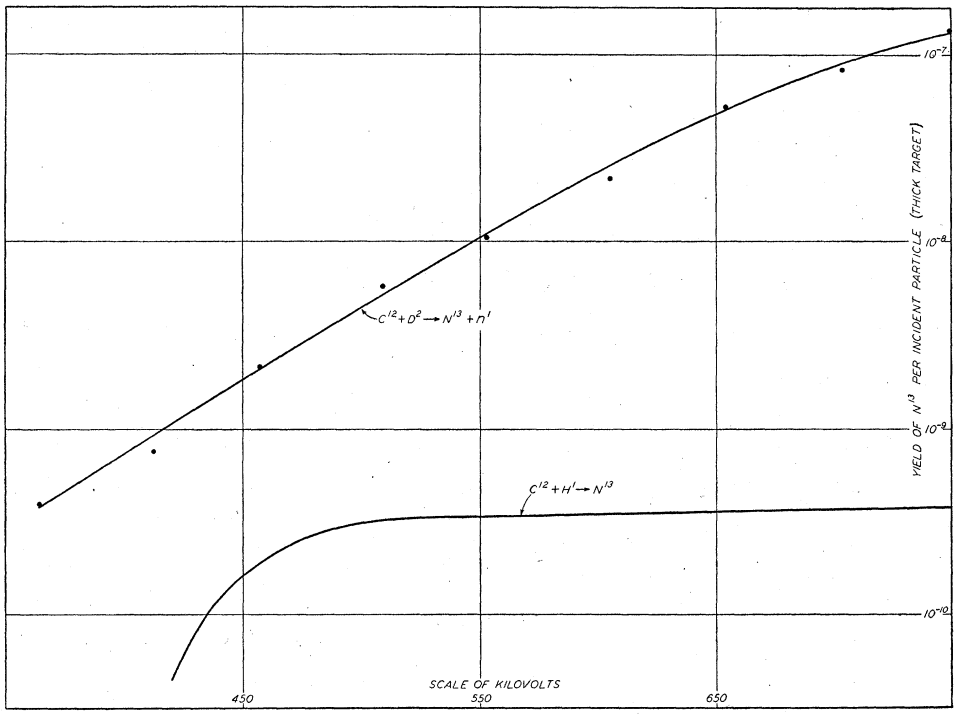

FIG. 2. Production of N^{13} by protons and deuterons.

measurement gave a reading of 452 kv for the resonance voltage. The difference of about three percent was attributed to changes in the voltmeter and a compensating change was accordingly made in the calibration. The carbon resonance was then measured and the lithium resonance measurement repeated, thus comparing the carbon resonance directly with the nearby lithium resonance. The voltage of the sharp rise in the carbon resonance, thus established, furnished a convenient check on the voltmeter for later experiments concerned with the possible existence of higher resonances in carbon. Also the lithium measurement was repeated from time to time as a further precaution. As the humidity decreased the calibration correction also decreased, coming to a final and steady value of approximately one percent at the end of October. The voltages of all points have been multiplied by a factor ranging from 0.97 to 0.99 chosen to place the lithium resonance at 440 kv.

To check the calibration of the voltmeter at higher voltages, the lithium gamma-rays were produced by bombardment with molecular hydrogen ions, giving an accurate point for calibration at 880 kv. This point, with the points at 440 and 0 kv, gave a new calibration curve which showed less departure from linearity (possibly due to heating of the resistors) than the original curve. As this calibration technique using the molecular beam had not been used previously, we repeated the measurements of Hafstad, Heydenburg, and Tuve on the fluorine resonances near 900 kv and found that the resonances occurred at voltages approximately one percent lower than the previously published values.[2] The present measurements give 880 kv and 930 kv for the doublet instead of 892 kv and 942 kv. These new values are almost certainly more accurate than the original values as they are directly compared with the 880-kv point established by the lithium resonance.

Results

Figure 1 shows the yield curve for a thick carbon target bombarded by protons. Every observation is shown on the curve with no adjustments except for the corrections to the voltage scale described above. The curve shows a single resonance of 30-kv half-width at 450 kv. The absolute yield, determined by the uranium comparison as described above, is indicated on the figure and is 3.5×10^{-10} at 560 kv.

Figure 2 shows the thick-target yield curve for the reaction $C^{12}+H^{2}\rightarrow N^{13}+n^{1}$ measured by the uranium comparison and relative values checked by a simultaneous count of the neutrons produced. The absolute yield indicated was determined by uranium comparison, and is less by a factor of 2.5 than the neutron yield from the same reaction measured by Amaldi, Hafstad, and Tuve.

Our attempt to observe the scattering of protons by carbon at the resonance voltage was unsuccessful, but in view of the theoretical importance of such an experiment we believe it advisable to include a brief account of the difficulties encountered. Anomalous scattering of protons is to be expected when a radiative capture resonance is too broad to be accounted for by the width of the gamma-ray level. The resonance-scattering anomaly must be observed with thin scatterers and at large angles (135–180°) to reduce the background of Coulomb scattering. Bethe[11] has pointed out that one would expect an increase by a factor of three in the backward scattering (180°) of protons by lithium at a proton energy of 440 kv where the lithium gamm-ray resonance occurs. However, it is practically impossible to obtain a thin film of lithium for the scatterer. For carbon, however, we might hope to observe a scattering anomaly at the energy of the broad proton-capture resonance as there are several carbon compounds in the gaseous state suitable for use as the scatterer. Acetylene, C_2H_2, is especially good, as the hydrogen would give no back scattering.

We have attempted to observe the large angle scattering of protons by C_2H_2 at the resonance voltage by use of the scattering chamber shown in Fig. 3. With the annular-ring construction we were able to obtain adequate counting rates even at the large angle selected, but in this chamber protons of less than 550-kv energy would not register reliably. At the resonance energy (450 kv) the range of the proton after collision and after passing through the window (1-mm stopping power) was too short to give reliable counts

[11] Bethe and Placzek, Phys. Rev. 51, 450–484 (1937).

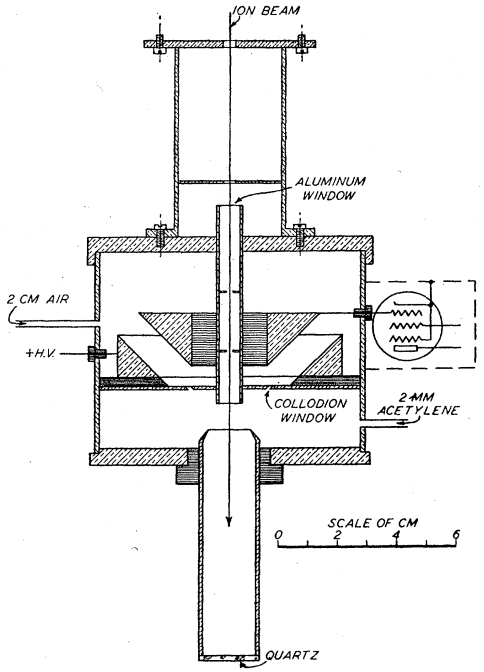

FIG. 3. Scattering chamber.

because of the large input capacity of the amplifier introduced by the annular collecting ring. We therefore concluded that this experiment requires the use of point counters for the detection of the scattered protons, although such a technique would encounter serious difficulties in obtaining adequate counting rates.

DISCUSSION OF RESULTS

The addition of the carbon resonance to the voltage scale established by the lithium and fluorine resonances furnishes another convenient voltage-calibration point. On account of its width the carbon resonance is somewhat less definite than the 440-kv lithium resonance, but this is perhaps balanced by the ease of preparation of carbon targets and their freedom from carbon contamination. The voltage of the carbon resonance is believed to be accurate to within two percent on an absolute scale, and somewhat better relative to the lithium and fluorine points. No indication of a fine structure of the carbon proton-resonance with respect to voltage appears in the present work.

The absolute yield of this proton reaction as given by the positron emission (based on our comparison with uranium) is approximately three times higher than that previously reported by Hafstad and Tuve, and gives a total reaction cross section of 3.35×10^{-24} volt cm^2. However, a more accurate method of determining the yield of N^{13} under proton bombardment is to accept the value for the yield of neutrons from carbon under deuteron bombardment published by Amaldi, Hafstad and Tuve as correct and equal to the yield of N^{13}. Then the yield of N^{13} from the proton reaction follows from the comparison of the number of positrons observed in the two reactions. This method gives a yield 2.5 times greater than the uranium comparison, corresponding to a total reaction cross section of 8.4×10^{-24} volt cm^2. The width of the gamma-ray level calculated from this value of the yield is then 0.6 volt, the lithium width being 4 volts.[12]

The yield of neutrons reported by Amaldi, Hafstad, and Tuve from the reaction $C^{12}+H^2\rightarrow N^{13}+n^1$ is believed to give a good value of the number of N^{13} atoms produced. Their method of determining neutron yields has been checked for the neutrons from deuterium bombarded by deuterium using an entirely different method, with agreement within 15 percent.[13] Moreover, there are no other reactions which could contribute appreciably to the neutron yield from the carbon. The reaction $C^{12}+H^2\rightarrow C^{12}+H^1+n^1$ is endothermic by 2.2 Mev and the reaction $C^{13}+H^2\rightarrow N^{14}+n^1$ is known to contribute only one percent to the number of neutrons observed.[14] The number of positrons observed from the reactions $C^{12}+H^2\rightarrow N^{13}+n^1$ as determined by the uranium comparison described above is less than the number of neutrons by a factor of 2.5. This discrepancy must be attributed either to estimating the number of positrons to be only 40 percent of the true number or to the conversion of N^{13} to C^{13} by K electron capture. In view of the errors involved in determining the number of positrons emitted, it would be unwise to offer this as definite evidence for K electron capture.

[12] Bethe, Rev. Mod. Phys. **9**, 207 (1937).
[13] Ladenburg and Kanner, Phys. Rev. **52**, 911–919 (1937).
[14] Bonner and Brubaker, Phys. Rev. **50**, 308–314 (1936).