

## Nitrogen 12\*

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$N^{12}$  is shown to have a half-life of  $12.5 \pm 1$  milliseconds, and a positron upper limit of  $16.6 \pm 0.2$  Mev. It is produced by the reaction  $C^{12}(p,n)N^{12}$ , and has a threshold proton energy of 20.0 Mev. This indicates that  $N^{12}$  is within about 200 kev of being unstable against proton emission. The mass of  $N^{12}$  is  $12.0228 \pm 0.00015$ , and the beta-transition is allowed.

### 1. INTRODUCTION

A RADIOACTIVE substance with a very short half-life was observed when high energy protons from a linear accelerator were incident on carbon. This activity has been studied in some detail and has been shown to be due to  $N^{12}$ . It is a rather interesting substance, in that it has the shortest lifetime of any known beta-emitter, and its positrons are more than three times as energetic as those from any other previously known radioactive isotope. The mass of  $N^{12}$  has been investigated theoretically by Barkas,<sup>1</sup> who concluded that its stability against proton emission is borderline.  $N^{12}$  is the third identified member of the new radioactive series with  $A = 2(Z - 1)$ .  $C^{10}$  and  $O^{14}$  have been observed by Sherr, Muether, and White.<sup>2</sup>

All of the radioactive measurements on  $N^{12}$  were made with the carbon target in its bombarded position. The normal time between beam pulses on the linear accelerator is 67 milliseconds, which is 5.3 half-lives. Therefore, the activity decays almost to background between pulses, and fifteen new samples of  $N^{12}$  are made each second. After the first experiment with a single counter, all later work was done with a pair of trays of Geiger counters, which were connected in coincidence. The counting circuit was sensitive for only 1/120 second, almost immediately after the proton beam was turned off. This "gating" of the circuit helped to reduce the background, and, in addition, a 1 mm thick piece of copper was placed between the counter trays. This absorber eliminated  $\gamma$ -ray coincidences from the  $C^{11}$ , which is also formed in the carbon target by high energy protons.<sup>3</sup> The background was always negligible, and, in fact, was largely due to cosmic rays.  $N^{12}$  samples with an activity of the order of one millicurie may be produced with the full beam current ( $1\mu$  amp. peak;  $4 \times 10^{-9}$  amp. average), but all measurements were done at greatly reduced beam intensities, to avoid saturation of the counters.

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<sup>1</sup> W. H. Barkas, Phys. Rev. **55**, 691 (1939).

<sup>2</sup> R. Sherr, H. R. Muether, and M. G. White, Phys. Rev. **74**, 1239A (1948) and Phys. Rev. **75**, 282 (1949).

<sup>3</sup> W. W. Chupp and E. M. McMillan, Phys. Rev. **72**, 873 (1947); E. M. McMillan and R. D. Miller, Phys. Rev. **73**, 80 (1948); W. K. H. Panofsky and R. Phillips, Phys. Rev. **74**, 1732 (1948).

### 2. HALF-LIFE MEASUREMENT

The half-life was measured with the aid of the gate circuit previously described. The repetition rate of the linear accelerator was cut from 15 cycles per second to 7.5 cycles per second, in order that the decay curve could be carried out to greater times. The width of the gate, or sensitive time of the counting circuit, was kept fixed at 0.008 second, and its delay time after the end of the proton pulse was varied by steps of 1/60 second, as measured on an oscilloscope. The number of positrons recorded through the gate for a given number of incident protons was measured as a function of the gate delay. The integration of the proton current was carried out by counting the  $C^{11}$  activity induced in a 0.001" polystyrene foil, through which the protons passed before striking the carbon target.

The half-life was measured several times in a period of several weeks, with different experimental arrangements, and the various values agreed within experimental error. Great care had to be taken to avoid overloading of the counters since, although the average counting rates appear to be

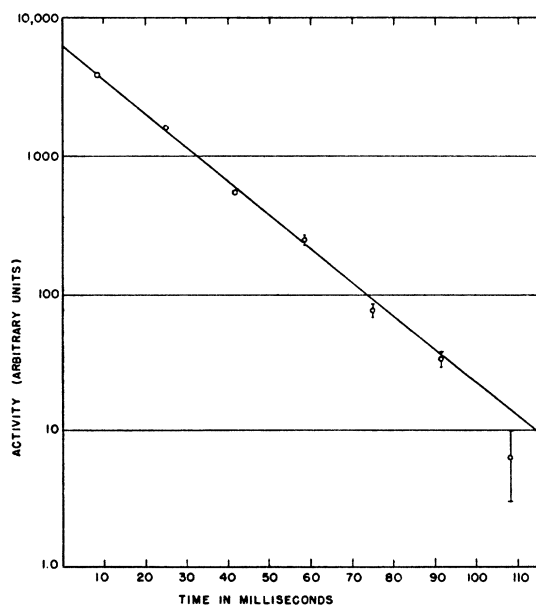


FIG. 1.  $N^{12}$  decay curve. Half-life =  $0.0125 \pm 0.001$  sec.

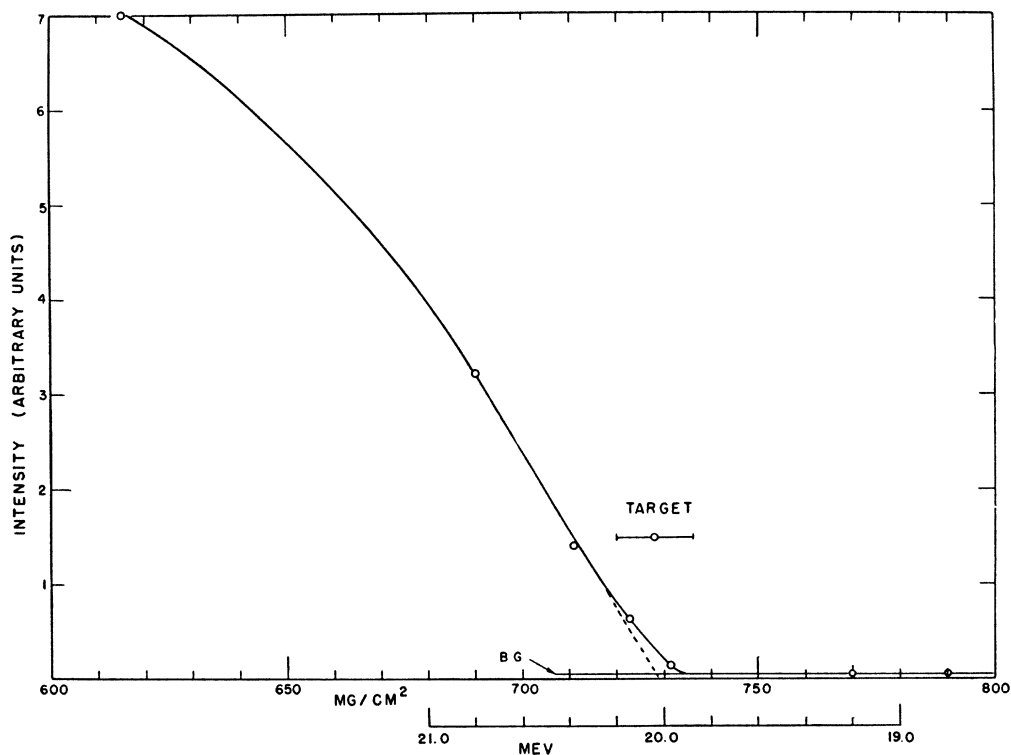


FIG. 2.  $N^{12}$  thin target excitation curve. Target 17 mg/cm<sup>2</sup> polystyrene. Initial proton energy = 32 Mev.  $C^{12}(p,n)N^{12}$ .  $N^{12} \rightarrow C^{12} + e^+$ .

relatively low, the instantaneous rates are very high because of the intermittent character of the operations. Figure 1 shows a typical decay curve over a factor of approximately 1000 in intensity, which yields a value of  $0.0125 \pm 0.001$  second for the half-life.

### 3. EXCITATION CURVE

Panofsky and Phillips have determined the excitation curve for the production of  $C^{11}$  by the bombardment of  $C^{12}$  by fast protons. The theoretical threshold of the  $C^{12}(p,pn)C^{11}$  reaction may be calculated from the known masses of the reacting nuclei, and is 20.2 Mev. The fact that  $C^{11}$  activity was observed at lower energies was interpreted as evidence for the  $C^{12}(p,d)C^{11}$  reaction. It may easily be shown that if  $N^{12}$  is stable against proton emission to form  $C^{11}$ , its  $p,n$  threshold on  $C^{12}$  must be lower than 20.2 Mev, the  $(p,pn)$  threshold. The binding energy of the last proton in  $N^{12}$  is equal to 12/13 of the difference between the  $p,pn$ , and the  $p,n$  thresholds.

The first excitation curves for the 12 millisecond period appeared to have thresholds in the neighborhood of 21 Mev. Thick targets were used in these experiments, however, and the extrapolations to zero activity were more difficult than if thin targets were used. Since the reasonable identification of the activity as  $N^{12}$  depended critically on the precise

value of the proton energy threshold, a number of thin target excitation curves were measured. The data for Fig. 2 were taken with a target of polystyrene 0.005" thick. The aluminum absorbers were weighed, and the tabulated values of the absorber mass per unit area include the aluminum window of the accelerator, the air path between the window and target, and one-half the thickness of the target. The range energy curve is from the work of Smith.<sup>4</sup> There are a number of reasons to believe that it gives the proper energies in the 20–30 Mev region. Immediately after several of the determinations of the proton threshold, calibrated nuclear emulsion plates were exposed to the protons from the linear accelerator. The protons were found to have a range corresponding to  $32.0 \pm 0.1$  Mev, which is the energy one calculates from the dimensions and frequency of the accelerator. Dr. Panofsky has measured the lengths of scattered proton tracks in the same plates, where the original protons had the full accelerator energy and were scattered from protons at known angles. This is an excellent way of extending the range-energy curve for nuclear emulsion plates from the lower energy region, where it is known accurately, to the range of 32 Mev. One makes use of the relationship,

$$E_{\theta} = E_0 \cos^2 \theta.$$

<sup>4</sup> J. H. Smith, Phys. Rev. **71**, 32 (1947).

In addition to these checks of the range-energy curve, one may work differentially from the measured threshold of the  $C^{12}(p,d)C^{11}$  reaction, correcting for the Gamow factor of the escaping deuteron. All methods of determining the energy of a proton in the 20 Mev range give results which check to better than  $\pm 100$  kev.

The threshold of the  $C^{12}(p,n)N^{12}$  reaction is at 20.0 Mev. If one assumes that the excitation curve near the threshold for an infinitely thin target is a straight line meeting the horizontal axis at a finite angle, then the curve for a target of finite thickness should extrapolate to the axis at "the center of the target." It should be curved over a range of absorber thickness equal to that of the target. Within the accuracy of the data in Fig. 2, that is just the appearance of the excitation curve. The effects of straggling, and of the initial energy inhomogeneity of the 32 Mev beam do not show in the curve. There is at present no method available in our laboratory sensitive enough to detect the energy spread in the 32 Mev beam. From this work, and that of Panofsky and Phillips, it is known that the energy spread is less than 100 kev, or less than  $3 \times 10^{-3}$ .

The excitation curve was found to level off at a

value of 10 arbitrary units for energies above about 24 Mev. No physical significance should be attached to this observation, because recoil of the  $N^{12}$  from the thin target foil would be expected in this range. With the stacked foil technique, this effect is not important, but in this case, no compensating factors offset the recoil effect. In one run with a much thicker target, the excitation curve was observed to rise continuously from 20 to 32 Mev as it should if the recoil hypothesis is correct.

#### 4. ABSORPTION DATA

The absorption of the  $N^{12}$  positrons is shown in Fig. 3. The intensity was reduced by a factor of about  $10^3$ , and the end point is of the so-called inspection type, which is generally more reliable than end points based on comparisons with known spectra. Any comparison with other positron emitters would involve rather large extrapolations, and the only convenient high energy negative electron emitter which may be produced by the accelerator is  $Li^8$ , from the  $Be^9(p,2p)$  reaction. Since this isotope has a complex spectrum, it would not be suitable for use with the Feather comparison method. The upper limit is calculated from Feather's formula, as corrected by Glendenin and Coryell,<sup>5</sup>

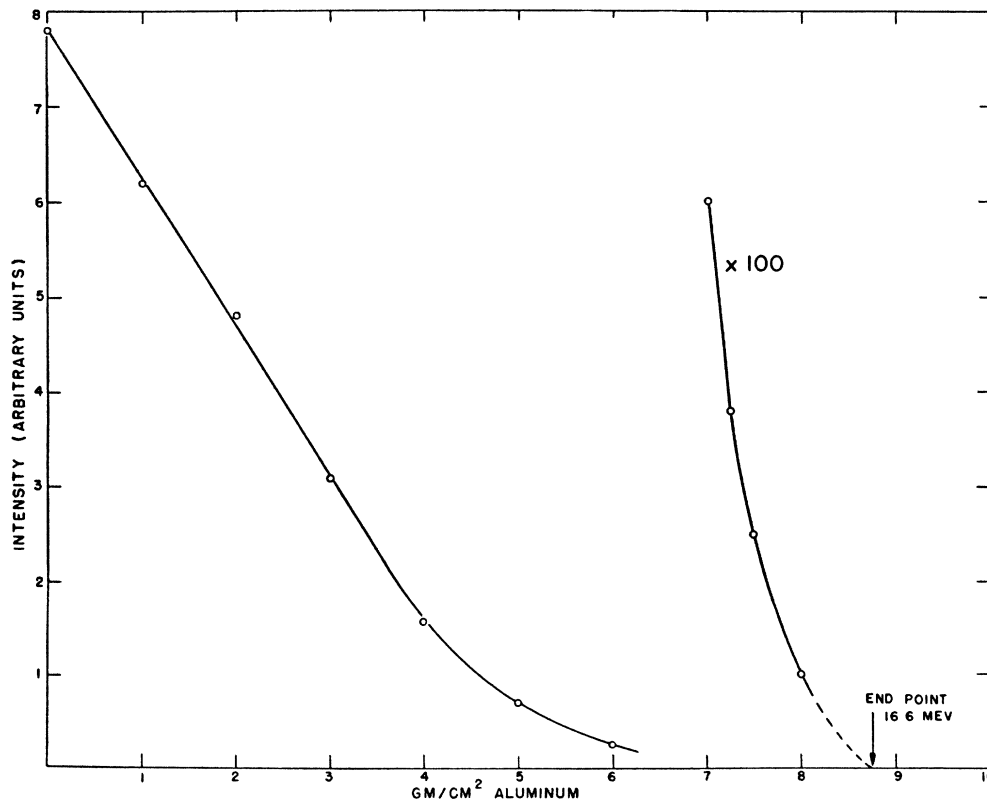


FIG. 3. Absorption of  $N^{12}$  positrons (background subtracted).

<sup>5</sup> L. E. Glendenin and C. D. Coryell, MDDC 19 (1946).

and has the value 16.6 Mev. This is precisely the upper limit one calculates from the atomic masses, assuming a  $p,n$  threshold of 20.0 Mev.

To show that the electronic component consisted of positrons, a search was made for annihilation radiation. A  $\gamma$ -ray was found which decayed with a period of approximately  $10^{-2}$  second. That this  $\gamma$ -ray was not of nuclear origin was shown by surrounding a bare source of  $N^{12}$  with a heavy copper cylinder. Lead diaphragms were placed so that the counters could "see" only the region near the  $N^{12}$  source. When the cylinder was placed around the  $N^{12}$  sample, the  $\gamma$ -ray counting rate increased, which showed that formerly, the positrons were annihilated at more distant points, from which the annihilation quanta could not reach the counters.

### 5. IDENTIFICATION OF THE ACTIVITY

In the early stages of the work, before accurate half-life and threshold values were available, it was thought that the activity might be the 0.022 second period of  $B^{12}$ , formed in the reaction  $C^{13}(p,2p)B^{12}$ . Bombardment of samples enriched in  $C^{13}$  showed that the activity definitely arose from the  $C^{12}$ .\*\*

The identification of the activity comes from the following energetic evidence:

(a) The masses of  $N^{12}$ , as computed from the proton threshold and the positron endpoint are the same to within the error of the measurements, or about  $\pm 0.0002$  mass unit.

(b) It may be shown by a series of detailed calculations that on energetic ground, all isotopes with  $Z$  less than 8, and  $A$  less than 14, may be ruled out as being responsible for the 0.0125 second activity. From the positron energy, one may compute the mass of any supposed responsible isotope, which decays into a known isotope. Then, using the observed proton threshold, the mass of the excited  $N^{13}$  atom is calculated. It has been shown that the only two nuclei which come within five Mev of satisfying the energetic requirements are  $Li^4$  and  $B^8$ , which are each off by about 4 Mev. (These nuclei would be formed in  $(p,\alpha n)$  or  $(p,2\alpha n)$  reactions.) In all cases, there is not enough energy available to make the reactions go. It is not just that the numbers do not agree perfectly as they do in the  $N^{12}$  calculation; the reactions are energetically forbidden. For all other isotopes, which lie further from the stability curve, it is possible to show that they are even more easily disposed of on energetic grounds.

\*\* I am indebted to Dr. J. W. Otvos of the Shell Development Company for the loan of the enriched carbon samples.

### 6. DISCUSSION

The mass of  $N^{12}$  is  $12.0228 \pm 0.00015$  M.U., based on  $C^{12} = 12.0038$ . The beta-transition from  $N^{12}$  to  $C^{12}$  is allowed. As calculated from the formula given by Konopinski,<sup>6</sup> the ft. value for the transition is  $1.6 \times 10^4$ , as compared with  $1.4 \times 10^4$  for  $B^{12}$ , using the latest values for the upper limit of  $B^{12}$ .<sup>7</sup>

According to Professor E. Teller,<sup>8</sup> the existence of  $N^{12}$  may be of some importance in theories of the evolution of the light elements.

$N^{12}$  may be the most nearly ideal radioactive isotope for testing the neutrino theory. The maximum recoil energy of a  $C^{12}$  nucleus from the reaction of a neutrino is 13,200 electron volts. Such recoils should have a range of about 2 cm in a cloud chamber operated with pure water vapor at a few degrees centigrade. The  $N^{12}$  could be deposited on a charged wire in an atmosphere of some hydrocarbon and then rapidly transported through a small hole into the low pressure cloud chamber. The direction of the recoil would be evident, and its energy could be measured by drop counting techniques. The drop counting technique could be calibrated by  $O^{16}$  recoils from neutrons incident on the chamber. The direction and energy of the positron could be photographed in an ordinary cloud chamber plus magnetic field, placed adjacent to the small, low pressure recoil chamber. If the experiment could be done, one would have the direction and energy of recoil and electron, and could compute from momentum consideration, the direction and energy of the neutrino. This computed energy could then be compared with the energy calculated from the positron energy and the upper limit. If the two values always agreed, one would gain still more confidence in the neutrino theory.

### ACKNOWLEDGMENTS

I am indebted to Mr. Donald Cone for constructing the electronic gate circuit, to Dr. Hugh Bradner for help with some of the measurements, and to the linear accelerator crew for the bombardments.

<sup>6</sup> E. J. Konopinski, Rev. Mod. Phys. 15, 209 (1943).

<sup>7</sup> W. F. Harnyak, C. B. Dougherty, and T. Lauritsen, Phys. Rev. 74, 1727 (1948).

<sup>8</sup> Private communication.