Ionization Chamber Study of the Disintegration of He^3 and N^{14} by Thermal Neutrons*

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The total ionization produced by the reactions $He^3(n,p)$ and $N^{14}(n,p)$ in a gas mixture consisting mainly of argon was measured by means of a cylindrical ionization chamber operated under conditions of electron collection. High resolution was achieved by suitable choice of chamber geometry and by use of a low noise level amplifier. The total ionization charges collected from polonium alpha-particles, from the disintegration products of $He^3(n,p)$ and from those of $N^{14}(n,p)$ are found to be in the ratio $1:0.145\pm0.002:0.119\pm0.001$, respectively. Comparison with polonium alpha-particles on the assumption of a constant average energy per ion pair in argon then yields reaction energies of 766 ± 10 kev for $He^3(n,p)$ and 630 ± 6 kev for $N^{14}(n,p)$. The difference between these energies agrees well with the difference between the end points of the beta-ray spectra of C^{14} and H^3. This agreement supports the assumption of a proportionality between energy and ionization in argon independent of particle type. A value of 785 ± 6 kev is computed for the neutron-hydrogen mass difference from these data.

INTRODUCTION

A PRECISE measurement of the energy evolved in (n,p) reactions is of interest in view of the fact that the initial and final nuclei involved in such reactions are neighboring isobars. The reaction energy when combined with the neutron-hydrogen mass difference will therefore yield a value for the difference between the masses of the two isobars, a quantity which may also be determined from the radioactivity of the final nucleus. Thus, consider a (n,p) reaction involving an initial nucleus of atomic number Z and mass number A:

$$Z^A+{}_0n^1\rightarrow(Z-1)^A+{}_1H^1+Q_{np}.$$

The reaction will proceed with slow neutrons if $Q_{np}>0$. On the other hand, if $Q_{np}<{}_0n^1-{}_1H^1$, the final nucleus is unstable and decays into the initial nucleus by negative electron emission:

$$(Z-1)^A\rightarrow Z^A+e^-+E_\beta+\nu.$$

If we consider atomic masses, we then have two possible expressions for the mass difference of the two isobars:

$$(Z-1)^A-Z^A=({}_0n^1-{}_1H^1)-Q_{np},$$
$$(Z-1)^A-Z^A=E_\beta+\nu.$$

Here E_β is the *actual* maximum energy of the electrons emitted by the final nucleus and ν is the mass of the neutrino.

In principle, therefore, it is possible to obtain an upper limit on the neutrino mass by use of a reaction cycle which involves the (n,p) reaction energy, the neutron-hydrogen mass difference and the maximum energy E_β of the beta-rays. On the other hand, by assuming that the end point E_{ext} of the beta-ray spectrum of the product nucleus as obtained by extrapolation on the basis of the Fermi theory gives the mass difference of the isobars (independent of the neutrino mass), that is, if $E_{\text{ext}}=E_\beta+\nu$, one may calculate the neutron-hydrogen mass difference by adding E_{ext} to

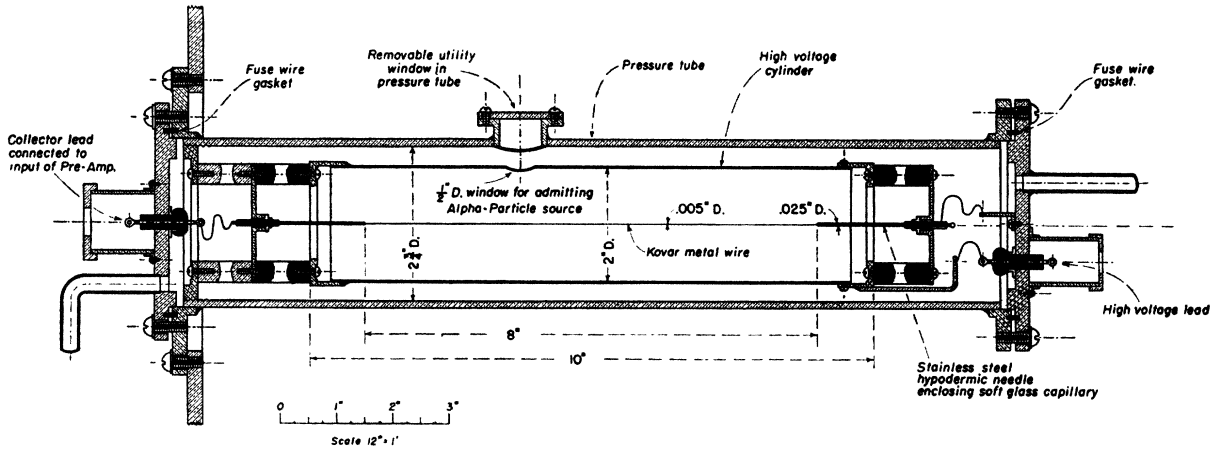

FIG. 1. Cylindrical chamber used for the study of slow neutron reactions.

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the (n,p) reaction energy: ${}_0n^1-{}_1H^1=E_{\text{ext}}+Q_{np}$. The second one of these two alternate procedures was regarded here as the best application of the data obtained.

Previous studies of slow neutron (n,p) reactions have in general fallen into two classes: Cloud-chamber range measurements and total ionization determinations. Despite the precision that can be attained in the measurement of the range of the emitted protons,[1-3] the interpretation of the resulting range values is handicapped by the uncertainty existing in the proton range-energy curve in the low energy region. The total ionization determinations, on the other hand, have been handicapped by the comparatively small energy evolved in slow neutron (n,p) reactions; as a consequence, amplifier noise proved to be a serious limitation on the energy resolution that could be attained, particularly in experiments performed under conditions of total ion collection. The size of the noise level in these cases is caused by the necessity of operating the amplifier with time constants which are long compared to the collection time of the ions; as a consequence, a great deal of low frequency noise (microphonics, line frequency hum, and thermal noise) is present. It is experimentally very difficult to reduce this type of noise below a level which represents an appreciable fraction of the (n,p) reaction energy. A more fundamental limitation on the interpretation of the ionization chamber data has been the absence of any precise knowledge regarding the relationship between energy and total ionization.

We have attempted to overcome the effect of amplifier noise by operating an ionization chamber with electron collection in a gas mixture having high electron drift velocity and by choosing the time constants of the associated amplifier so as to achieve as low a noise level as practicable. A study of the various factors that contribute to the total noise level made it possible to reduce this level to a value of the order of one percent of the energies to be measured. The difficulty introduced by the lack of a reliable energy-ionization relation was minimized to some extent by using argon (in which this relation has been studied more thoroughly than in most other gases) as the principal stopping gas in the chamber. In addition, the two reactions were observed simultaneously so that a cross-check on the validity of the method of calibration could be obtained.

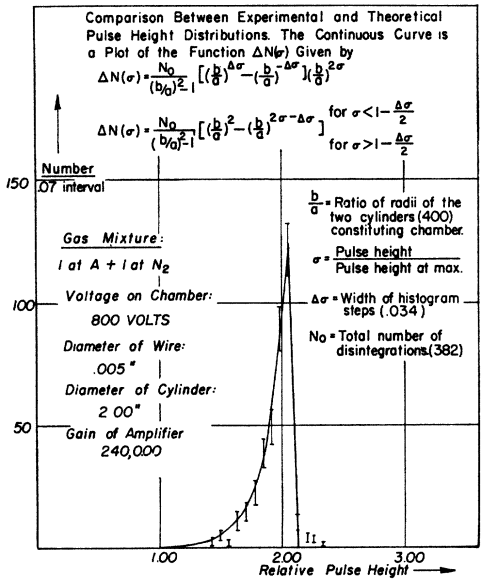

FIG. 2. $N^{14}(n,p)$ reaction. Comparison of experimental pulse-height distribution with the theoretical distribution derived by considering the effect of electron collection on an isotropic distribution of point charges throughout the volume of a cylindrical chamber of the dimensions used.

EXPERIMENTAL METHOD

The cylindrical ionization chamber used in the experiment is shown in Fig. 1. To be noted particularly are the use of guard cylinders to prevent collection of the ionization electrons formed outside the region of cylindrical symmetry, and the large ratio between the diameters of the two cylinders that constitute the chamber.

The reason for using a large ratio of radii is that do to the use of electron collection, the voltage signal induced by the motion of the free electrons from their point of liberation to the collector electrode is in general smaller than the signal which would be induced by the motion of the entire charge, including the positive ions. As a matter of fact, the size of the induced signal is proportional to the fraction of the total potential difference (between the electrodes) which is traversed by the electrons.[4] Thus, corresponding to a known initial geometrical distribution of charges, one obtains a distribution of voltage signals of various sizes. For an isotropic distribution of very short disintegration tracks throughout the volume of a cylindrical chamber, the differential distribution in pulse heights can be computed simply:[5]

$$dN/d\sigma=\frac{2N_0\log(b/a)}{(b/a)^2-1}(b/a)^{2\sigma} \text{ for } 0<\sigma<1,$$

$$dN/d\sigma=0 \text{ for } \sigma>1.$$

Here σ is the ratio of the actual pulse height to the pulse height at the maximum of the distribution curve, N_0 is the total number of disintegrations, and a and b are the radii of the two cylinders constituting the chamber. This distribution is sharply peaked at $\sigma=1$ if $b\gg a$; its width at half-maximum is given by

[1] J. K. Boggild, Kgl. Danske Vid. Sels. Math.-fys. Medd. **23**, 22 (1945).
[2] D. J. Hughes and C. Eggler, Phys. Rev. **73**, 809 (1948).
[3] Cornog, Franzen, and Stephens, Phys. Rev. **74**, 1 (1948).
[4] See, for example, Bridge, Hazen, Rossi, and Williams, Phys. Rev. **74**, 1084 (1948).
[5] P. G. Koontz and T. A. Hall, Rev. Sci. Inst. **18**, 643 (1947).

$[(\frac{1}{2})\log 2]/\log(b/a)$. To make this width as narrow as possible, it is advantageous to use as large a ratio b/a of the radii as possible.

The use of guard cylinders, on the other hand, makes it possible to collect electrons only from those disintegration tracks which are formed in the cylindrically symmetric electric field, and to exclude those which are formed in the distorted field at the ends of the cylinder. As a consequence, the experimentally observed pulse-height distribution closely resembles the theoretical distribution defined above with the modifications that are introduced by the finite size (channel width) of the groups into which the pulses are arranged, and by the presence of wall effect, amplifier noise, straggling in ionization and similar factors. (The close agreement that can nevertheless be achieved between experimental and theoretical distributions is illustrated by Fig. 2.)

Not shown is a retractable thin polonium alpha-particle source on a platinum foil which was attached, by means of a glass rod and a sylphon bellows, to the outside wall of the chamber and which could be positioned in the plane of the high voltage cylinder. When not in use, the source could be withdrawn to a position where none of the alpha-particles emitted by it were able to reach the sensitive volume of the chamber provided the stopping power of the gas filling exceeded one and one-half times that of air at N.T.P. The source itself was made by deposition from 0.1N HCl solution under hydrogen, as described by Erbacher.[6]

The amplifier used with the chamber was a modified version of the Los Alamos Model 100 amplifier designed by Sands.[7] The modifications consisted of operating the first tube of the preamplifier as a triode, of moving the low frequency limiting RC network to a location between the two feed-back loops of the main amplifier (the time constant of this network was 5 μsec.), and of by-passing the plate resistance of the signal inverting stage (last stage of first feed-back loop of main amplifier) with a condenser of such a magnitude as to make the time constant of this network 7.5 μsec., corresponding to a "rise time" of 17 μsec. This combination of time constants was found experimentally to result in the largest attainable signal-to-noise ratio. In the examination of the effect of different combinations of time constants on the signal-to-noise ratio, we were guided to a large extent by the considerations advanced by Elmore.[8] It was found, however, that various low frequency sources of noise (particularly microphonics) made it difficult to approach the optimum combination of time constants predicted by Elmore. Moreover, it was regarded desirable to keep the duration of the pulses observed at the output of the amplifier within a certain limit to prevent excessive "pile-up" of gamma-ray pulses due to gamma-rays emitted by the radium-beryllium neutron source.

Other modifications of the amplifier consisted of careful selection of the input tube of the preamplifier, non-microphonic mounting, selection of other components at critical stages, and use of coupling time constants between stages at least 200 times as large as the time constant of the low frequency limiting network. The last precaution mentioned made it possible to eliminate any evidence of "overshoot." The input tube was operated with floating grid under reduced electrode voltages; the leakage resistance between the input grid and ground was estimated to be 10^{12} ohms by observation of the time constant with which the capacity between the collector wire and the high voltage cylinder charged up when connecting the cylinder to a source of high potential. Under these conditions, the over-all gain of the amplifier was 240,000 for artificial pulses having a "rise time" an order of magnitude shorter than the time constants of the amplifier, and the root-mean-square noise voltage at the input to the amplifier was 2.3 μv, corresponding to 8 kev of ionization for the input capacity used. (This capacity was estimated to be 20 $\mu\mu$f under operating conditions; the measured capacity with input tube turned off was 16 $\mu\mu$f.)

An illustration of the resolution achieved under these conditions is provided by Fig. 2 in which the distribution in size of 382 individual pulses attributed to $N^{14}(n,p)$ is compared with the theoretical distribution derived by considering the effect of electron collection on an isotropic distribution of point charges distributed throughout the volume of a cylindrical chamber of the dimensions used. The theoretical distribution has been normalized to correspond to the same total number of disintegrations as were actually observed, and the effect of the finite width of the individual groups of the histogram has been taken into account. It is evident that the additional spread in pulse height caused by amplifier noise is small; some straggling in ionization undoubtedly also contributes to the observed spread.

Pulses were observed by photographing the face of an oscilloscope tube; for this purpose the shutter of a Sept camera was held open manually until a signal appeared on the face of a coupled monitoring oscilloscope. At this point, the film was advanced by a single frame. Thus signals occurring at a rate as slow as three per minute could be recorded. The linearity of the over-all system was tested by means of a precision pulser and an electronic discriminator.

The gases used in the chamber were carefully purified by use of the purification system illustrated in Fig. 3. Mixtures of argon and nitrogen could be circulated through the calcium oven shown which was maintained at a temperature of 200°C. At this temperature, the calcium does not react with nitrogen, but is nevertheless very effective in removing contaminants that may give rise to electron capture. The calcium itself was outgassed at 400°C for 24 hr. in a vacuum of the order of

[6] O. Erbacher, Zeits. f. physik Chemie **A156**, 142 (1931).
[7] M. Sands, LADC 82.
[8] W. C. Elmore, Nucleonics **2**, No. 3, 16 (1948).

5×10^{-6} mm Hg before use. The progress of the purification could be checked periodically by taking integral bias curves of polonium alpha-particle pulses as a function of applied potential. For the gas mixture used in the helium-nitrogen-argon mixture experiment, the shift that occurred in the position of the maximum of the pulse-height distribution amounted to one percent of the maximum pulse height as the applied potential was increased from 100 volts to 1000 volts after purification had been completed.

The neutrons originated from a 200 mC radium-beryllium source surrounded by a cylinder of paraffin which was placed below the chamber in such a way that the flux of slow neutrons traversed the chamber along its axis. The thickness of paraffin used was not less than 5 cm on all sides; under these conditions, there was an observable background of fast neutron recoil pulses. However, even in the case of the experiment performed with a gas mixture that contained enriched helium in which rather energetic recoils occur, this background was not large enough to interfere with the observation of the monoenergetic disintegration pulses. Approximately 10 cm of lead were placed in the direct path between the source and the chamber in order to keep the background of gamma-ray pulses to a minimum.

RESULTS

The simultaneous observation of the reactions $He^3(n,p)$ and $N^{14}(n,p)$ was accomplished in a gas mixture consisting of two atmospheres argon, one-tenth atmosphere nitrogen, and one-half atmosphere helium which had been enriched by thermal diffusion at the University of Minnesota. The relative isotopic abundance of the two helium isotopes in the sample used was $He^3/He^4=4.9\times10^{-5}\pm25$ percent. The sample was supplied through the kindness of Professor A. O. C. Nier of the University of Minnesota.

Figure 4 shows the resulting pulse-height distributions as recorded photographically. The mixture of the enriched helium with nitrogen thus gives rise to a double peak with two well-resolved components attributed to $He^3(n,p)$ and $N^{14}(n,p)$, respectively. The relative amounts of the isotopes N^{14} and He^3 in the chamber were originally chosen in such a way that, on the basis of the thermal cross sections published by Coon *et al.*,[9] 7500×10^{-24} cm^2 for $He^3(n,p)$ and 1.75×10^{-24} cm^2 for $N^{14}(n,p)$, the ratio of the disintegration rate of $N^{14}(n,p)$ to that of $He^3(n,p)$ would be 2:1. This choice of the ratio of disintegration rates was dictated by a desire to avoid interference of the low energy tail of the pulse-height distribution due to $He^3(n,p)$ with the distribution due to $N^{14}(n,p)$. The actual ratio of disintegration rates was nearly 3:1 and is in approximate accord with recently published values of the cross sections for the two reactions,[10–12] averaging about 5000×10^{-24} cm^2 for $He^3(n,p)$ and 1.76×10^{-24} cm^2 for $N^{14}(n,p)$. No attempt was made here to measure the relative cross sections for the two reactions because of the uncertainty attached to the isotopic abundance of the He^3 isotope in the gas mixture.

The distribution in pulse heights obtained from a polonium alpha-particle source in the same gas mixture is shown in Fig. 5. For the observation of the alpha-particle pulses, the gain of the amplifier was diminished by a known amount, and the trigger level of the oscilloscope sweep circuit was set so that only alpha-particle signals were recorded. A comparison of the three

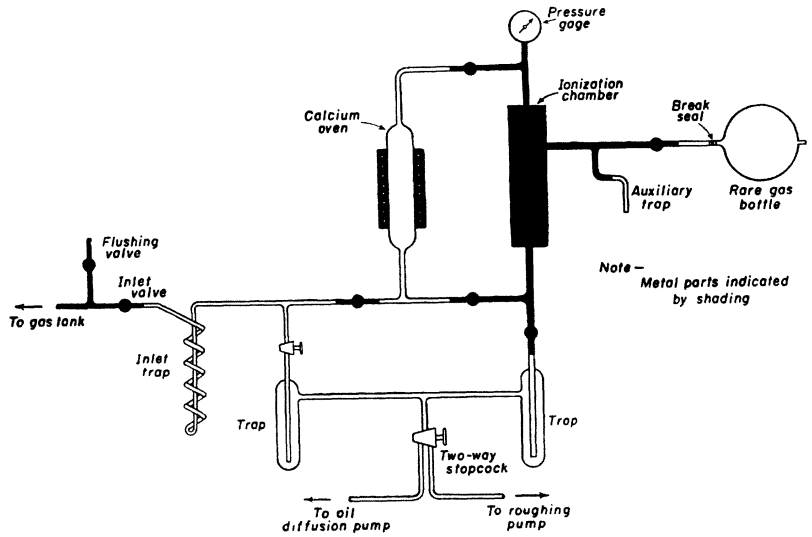

FIG. 3. Schematic diagram of gas-flow and pumping system.

[9] Coon, Goldblatt, Nobles, and Robinson, AECD 2190.
[10] J. H. Coon and R. A. Nobles, Phys. Rev. **75**, 1358 (1949).
[11] L. O. P. King and L. Goldstein, Phys. Rev. **75**, 1366 (1949).
[12] Batchelor, Epstein, Flowers and Whittaker, Nature **163**, 211 (1949).

pulse-height distribution curves reveals the characteristic features previously mentioned that typify the operation of a cylindrical chamber with electron collection. We observe that there is a gradual spread in the low energy direction for the (n,p) reactions, as one would expect for an isotropic distribution of short disintegration tracks throughout the volume of the chamber. The distribution is narrower for the alpha-particles due to the localization of the source on the wall of the ionization chamber. On the high energy side, all distributions fall off sharply in a manner which is determined chiefly by the noise of the amplifier. The sharpness of the location of the high energy edge enables one to deduce the total ionization corresponding to each of the distributions with precision.

A comparison of the three distributions on this basis gives a ratio of $1:0.145\pm0.002:0.119\pm0.001$ for the total ionizations produced in the mixture by polonium alpha-particles, the disintegration products of $He^3(n,p)$ and those of $N^{14}(n,p)$, respectively. The uncertainties indicated for the values of these ratios are estimates of the precision with which the position of the peaks of the pulse-height distributions can be located in view of statistical factors and the finite slope of the distribution curves on the high energy side. Deviations from linearity of the amplifier and recording system are of the order of one-tenth as large as the uncertainties just mentioned.

If we assume that the proportionality between energy and ionization in argon which has been observed by Jesse[13] (although disputed to some extent by the observations of Cranshaw and Harvey[14]) extends to the energy region considered here, and if the effect of the presence of one-tenth atmosphere nitrogen (responsible for approximately 10 percent of the total stopping power of the mixture) on this relation is neglected, we obtain energy values of 766 ± 10 kev for $He^3(n,p)$ and 630 ± 6 kev for $N^{14}(n,p)$ by direct comparison with polonium alpha-particles. (Helium is assumed to be similar to argon in its ionization properties in accordance with the conclusions of Gray.[15]) The polonium alpha-particles were assumed to have an energy of 5298.4 kev.[16] (It should be emphasized that the indicated uncertainties in the Q-values for the (n,p) reactions do not reflect any possible systematic errors arising from the method of calibration.)

Several other gas mixtures containing nitrogen and argon in various proportions were also studied, and a systematic increase with nitrogen concentration of the ratio of the total ionization due to $N^{14}(n,p)$ to that due to polonium alpha-particles was observed. This increase

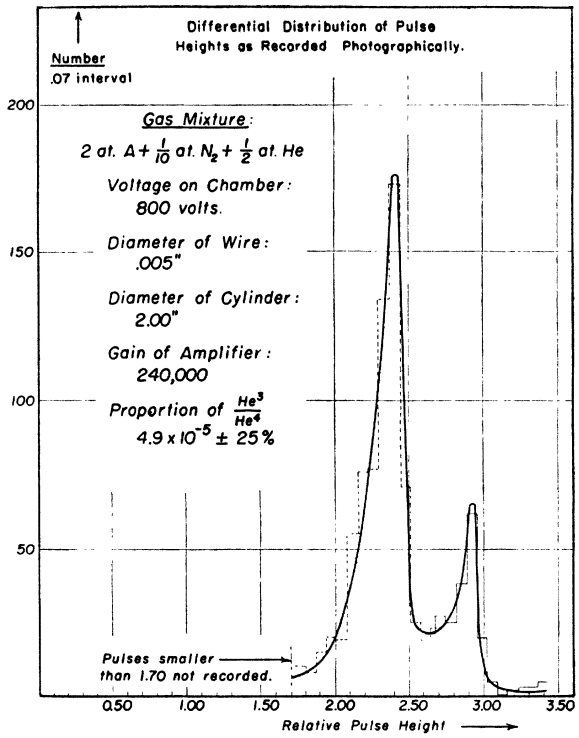

FIG. 4. Pulse-height distribution obtained from the reactions $He^3(n,p)$ and $N^{14}(n,p)$.

amounted to approximately three percent for pure nitrogen as compared to the helium-nitrogen-argon mixture. Thus the correction that would have to be applied to the energy values deduced from the total ionization observed in this mixture to account for the influence of the small concentration of nitrogen present amounts to -0.3 percent. This is negligibly small compared to other uncertainties inherent in the measurement.[17] The cause of the increase in total ionization with nitrogen concentration is not known, but is not necessarily in conflict with two other recent observations[18,19] in which a decrease of total ionization with energy in nitrogen as compared to argon was observed at the energy of the $B^{10}(n,\alpha)$ reaction and the energy of samarium alpha-particles. In the first of these experiments, the total ionization was measured in air, rather than in pure nitrogen. Under such conditions, there is no free electron mobility and saturation conditions are essentially different from those prevailing in purified nitrogen as used here.

DISCUSSION

The difference between the Q-values for $He^3(n,p)$ and $N^{14}(n,p)$ obtained by assuming simple proportionality

[13] (a) W. P. Jesse and H. Forstat, Phys. Rev. **74**, 1259(A) (1948); (b) W. P. Jesse and J Sadauskis, Phys. Rev. **75**, 1110 (1949).
[14] T. E. Cranshaw and J. A. Harvey, Can. J. Research **26**, 243 (1948).
[15] L. H. Gray, Proc. Camb. Phil. Soc. **40**, 72 (1944).
[16] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 (1938).
[17] For this reason, the correction of -2 kev that was applied to the energy values for $He^3(n,p)$ and $N^{14}(n,p)$ in a preliminary account of the experiment described here [Franzen, Halpern, and Stephens, Phys. Rev. **76**, 317 (1949)] has not been applied to the values given above.
[18] See reference 13(b).
[19] Stebler, Huber, and Bichsel, Helv. Phys. Acta **22**, 362 (1949).

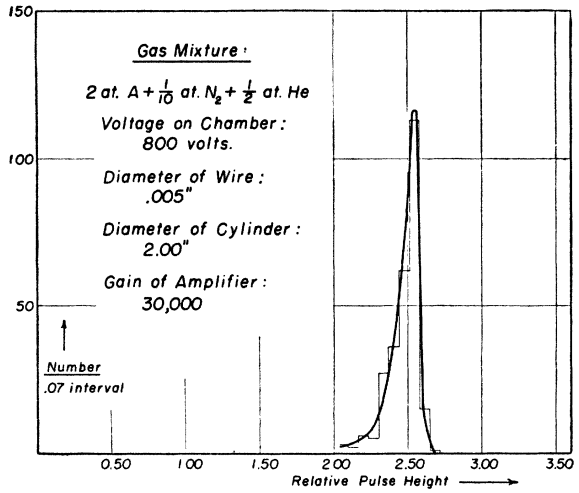

FIG. 5. Pulse-height distribution obtained from a wall source of polonium alpha-particles. The conditions were identical with those used for $He^3(n,p)$ and $N^{14}(n,p)$ experiments.

between energy and total ionization in argon agrees well with the value that one would expect from the end-point energies of the beta-ray spectra of H^3, 18±1 kev,[20,21] and C^{14}, 156±1 kev.[22] This agreement lends support to the method of calibration.

The energies obtained are in good agreement with recent observations of the thresholds for the inverse (p,n) reactions, 620±9 kev for $C^{14}(p,n)$,[23] and 764±1 kev for $H^3(p,n)$.[24] By adding to the measured Q-values the extrapolated end-point energies of the beta-ray spectra of C^{14} and H^3, we obtain an average neutron-hydrogen mass difference of 785±6 kev. This value is definitely smaller than the value obtained by Bell and Elliott (804±5 kev) from study of the gamma-rays emitted in $H(n,\gamma)$[25] and the HH−D mass doublet.[26,*] However, our value agrees well with the value for the neutron-hydrogen mass difference computed by Fowler and collaborators[27] (789±6 kev) from a different transmutation cycle.

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[20] G. C. Hanna and B. Pontecorvo, Phys. Rev. **75**, 983 (1949).
[21] Curran, Angus, and Cockroft, Phil. Mag. **40**, 53 (1949); Phys. Rev. **76**, 853 (1949).
[22] Cook, Langer, and Price, Phys. Rev. **74**, 548 (1948).
[23] Shoupp, Jennings, and Sun, Phys. Rev. **75**, 1 (1949).
[24] Taschek, Jarvis, Argo, and Hemmendinger, Phys. Rev. **76**, 325 (1949).

[25] R. E. Bell and L. G. Elliott, Phys. Rev. **74**, 1552 (1948).
[26] W. R. Cohen and W. F. Hornyak, Phys. Rev. **72**, 1127 (1947).
* *Note added in proof:* A revised value for the energy of the gamma-rays emitted in $H(n,\gamma)$, as obtained from the latest work performed by Bell and Elliott, leads to a neutron-hydrogen mass difference of 799 kev when the value cited in reference 26 for the HH−D mass doublet is used. (This result was communicated to the authors by Dr. W. B. Lewis of Chalk River.) On the other hand, if this revised gamma-ray energy is combined with the new value for the HH−D mass doublet recently obtained by Nier [T. R. Roberts and A. O. C. Nier, Phys. Rev. **77**, 746 (1950)] one obtains a value of 789 kev for $n-H$, in substantial agreement with the results of the disintegration experiments.
[27] Tollestrup, Jenkins, Fowler, and Lauritsen, Phys. Rev. **75**, 1947 (1949).