AECD - 2207
(LADC - 532)

UNITED STATES ATOMIC ENERGY COMMISSION

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by

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Date of Manuscript: June 9, 1948
Date Declassified: August 9, 1948

Technical Information Branch, Oak Ridge, Tennessee
AEC, Oak Ridge, Tenn., 4-22-49--850-A1895

Printed in U.S.A.
PRICE 10 CENTS

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ABSTRACT

The isotopic abundance of He^3 in one sample each of "well" helium and "atmospheric" helium has been measured by detecting the $He^3(n,p)H^3$ disintegrations induced by thermal neutrons. The helium gas was put into a proportional counter, the disintegration rate compared to that with nitrogen in the counter, and the He^3 content deduced using the known ratio of the He^3 and N disintegration cross sections.

* * * * *

Analysis of He^3 concentrations is of importance because of the interest in He^3 for nuclear investigations in the light element region. In the present work measurements were made of the isotopic abundance of He^3 in two samples of natural helium: one from wells near Amarillo, Texas, and one from air reduction processing. The method of measurement is applicable to analysis of the He^3 content of enriched samples prepared by thermal diffusion methods or by the production of He^3 through the decay of H^3.

The presence of He^3 was detected by counting ionization pulses arising from the disintegration products of the reaction $He^3(n,p)H^3$ induced by thermal neutrons. The cross section for this reaction is about 7500 barns[1] which is sufficiently large to make it possible to detect the He^3 in natural samples in which the concentration is only about 10^{-7}. Data were also taken with nitrogen in the counter, in which case one detects the $N^{14}(n,p)C^{14}$ disintegrations. Since the protons from the He^3 and N reactions have very closely the same range, the wall effect corrections will be similar for the two cases. If counting is done on nitrogen and on helium with the same neutron flux, then the disintegration rates D taking place within the active volume of the counter will be related by:

$$\frac{D_N}{D_{He}} = \frac{\sigma_N n_N}{\sigma_{He^3} n_{He^3}} \qquad (1)$$

where σ is the disintegration cross section and n the number of atoms of the gas in the counter. If I is the relative concentration of He^3 in the helium and p is pressure, then the above ratio may be written

$$\frac{D_N}{D_{He}} = \frac{\sigma_N}{\sigma_{He^3}} \quad \frac{2p_N}{Ip_{He}} \qquad (2)$$

The ratio σ_N / σ_{He^3} has been measured[1] and we can therefore obtain I by determining the counting rates at measured pressures.

The ionization pulses were detected in a cylindrical proportional counter three inches in diameter, with a 0.005 inch diameter center wire and with active length of about 5.75 inches. Positive voltage was applied to the center wire and the pulse was taken off the center wire by condenser coupling. The voltage was adjusted to give a gas multiplication of the order of 20. A Los Alamos Model 100 pulse amplifier[2] was used with an 80 microsecond "clipping time". The output of the amplifier was fed into a 10 channel differential pulse height selector[3] which analyses the pulse height distribution by dividing a chosen range of pulse heights into 10 equal pulse height intervals and counting the number of pulses having heights within each interval.

An overall check on the operation of the detecting and counting apparatus was made by counting alpha particles emitted from a thin deposit of normal uranium on a foil mounted near the inner wall of the counter. This source was mounted on a rotating disc and could be removed from seeing the counting volume by rotating the counter. Figure 1 shows a pulse height distribution curve obtained for the two alpha-particle groups. Values of 4.20 and 4.76 Mev were taken for the alpha-particle energies and served as an energy standard in looking for the proton groups from the (n,p) reactions. The observed pulse height with helium or nitrogen in the counter corresponded roughly to the known disintegration energies of 0.74 and 0.60 Mev though no attempt was made toward an accuracy better than about 0.2 Mev in correlating energies by pulse height comparisons.

For counting the disintegrations the counter was placed in a cavity near the vertical axis of the 5 ft by 5 ft by 9 ft high graphite pile of the Los Alamos neutron standards laboratory. Radium-beryllium neutron sources totaling about 6 grams of radium were also placed in the graphite pile at a distance of 48 inches above the counter. Counts were taken in this geometric arrangement with four different gas fillings in the counter as listed in Table 1. The high pressures of "well" helium were measured on a calibrated Bourdon tube gauge with 0.5 psi scale divisions and the lower pressures were measured with a mercury manometer. It is seen from Table 1 that the helium filled counter operates at an exceptionally low voltage. Argon was added to the nitrogen to increase the stopping power in the counter without causing the excessively high operating voltage required by pure nitrogen at higher pressure. In the case of "well" helium a small amount of argon was added to improve the operating characteristics of the counter, since with highly purified helium it was possible to obtain a maximum gas multiplication of only about five. An increase in voltage in attempt to obtain multiplication greater than five had little effect on pulse height but rapidly increased the "noise" level as though an electric discharge were taking place in the counter.

Since the effect of a small N_2 impurity in the helium would not be distinguishable from the effect of He^3, highly purified helium was used and the gas was further purified by continuous flow over calcium turnings at about 300°C. This calcium purifier was attached directly to the counter in the manner of Klema and Barschall.[4] Its effectiveness in removing nitrogen was tested by adding air to the argon filled counter, and observing a drop in the neutron induced counting rate as the calcium removed the nitrogen. An amount of nitrogen equivalent in respect to slow neutron capture, to the He^3 in "well" helium was entirely removed, within the detection sensitivity, in one day's continuous operation of the purifier.

The counting with any particular gas filling was done alternately with and without a cadmium cover over the counter for several cycles to average out fluctuations. To obtain an approximate measure of the disintegration rate, the count with cadmium is then subtracted from the count without cadmium. In the case of N_2 (see Figure 2), in the region near the peak of the pulse height distribution curve, the counting rate without cadmium was about 800 times the rate with cadmium. The count with cadmium increases in the small pulse region principally because the counter is sensitive to gamma-ray induced pulses.

In the case of helium (see Figures 3 and 4) the count without cadmium is relatively high because of the presence of a few residual high energy neutrons getting to the region of the counter in spite of the large amount of graphite between the counter and the fast neutron source. The He^4 recoils from these fast neutrons are appreciable in number because of the tremendous abundance of He^4 as compared to He^3. Figure 3 also shows the cadmium difference curve obtained with 1.95 atmospheres

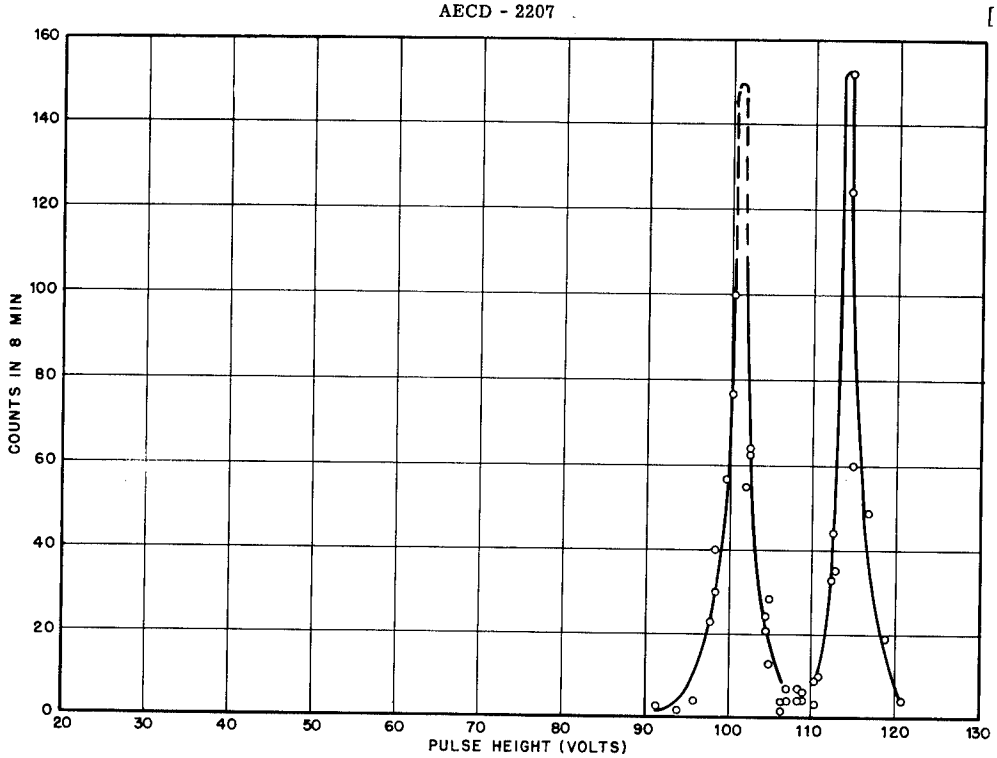

Figure 1. Differential pulse height distribution curve obtained with a thin deposit of normal uranium mounted on the inner wall of the counter. The two peaks correspond to the alpha-particle groups from U^{234} and U^{238}. 3.0 atm argon pressure.

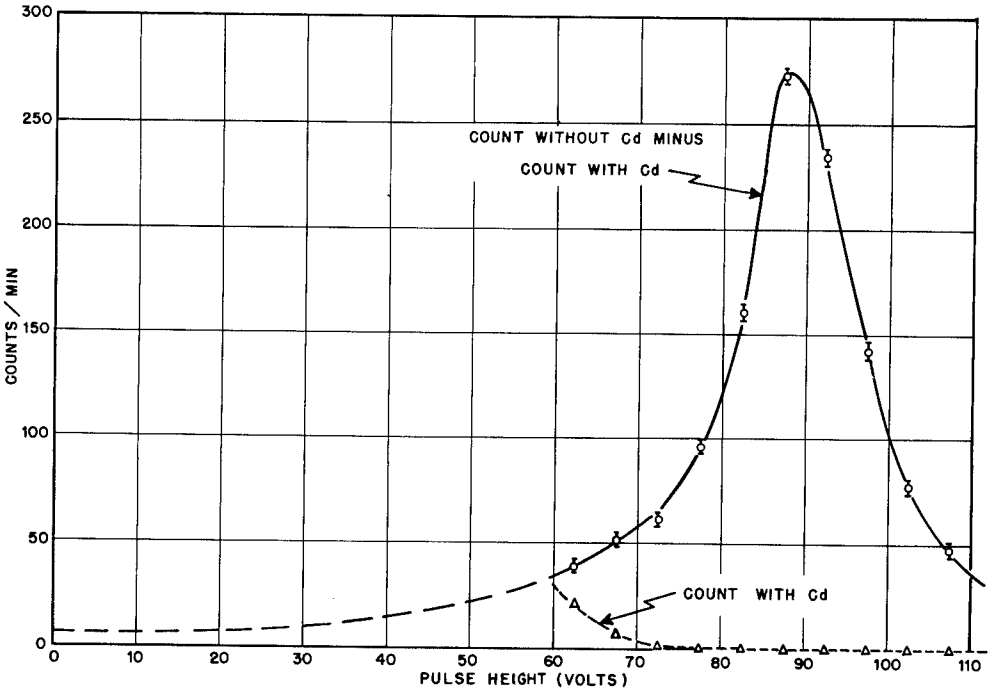

Figure 2. Pulse height distribution curves obtained by counting $N^{14}(n,p)C^{14}$ disintegrations. The plotted points represent the number of pulses in a 5 volt pulse height interval and the point is plotted at the center of the interval. The vertical lines indicate standard deviations computed by taking the square root of the total number of counts obtained and adjusting this to the counting rate.

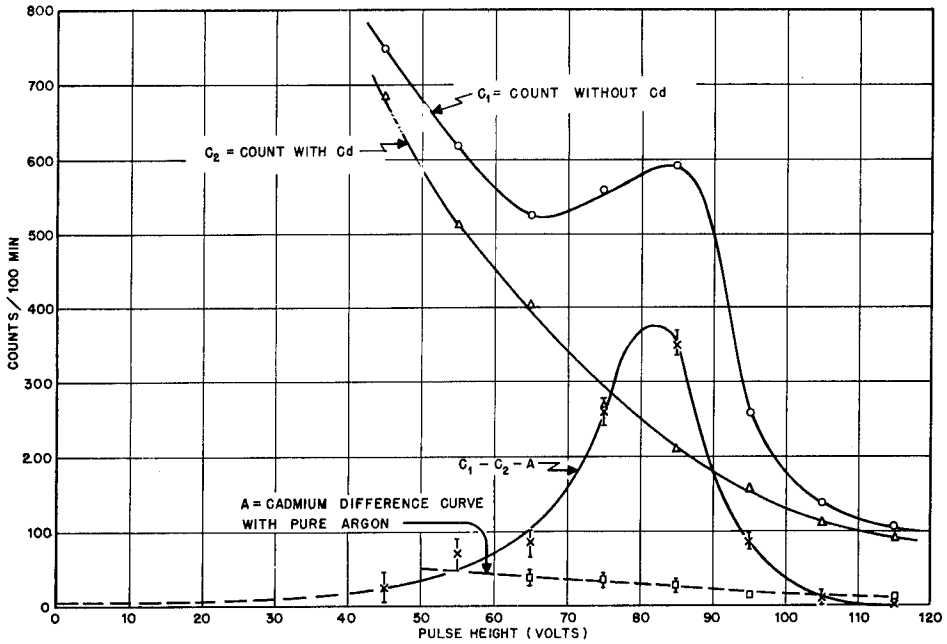

Figure 3. Pulse height distribution curves obtained with "well" helium. The plotted points represent the number of pulses in a 10 volt pulse height interval and the point is plotted at the center of the interval. The difference curve, $C_1 - C_2 - A$, corresponds to the number of $He^3(n,p)H^3$ disintegrations.

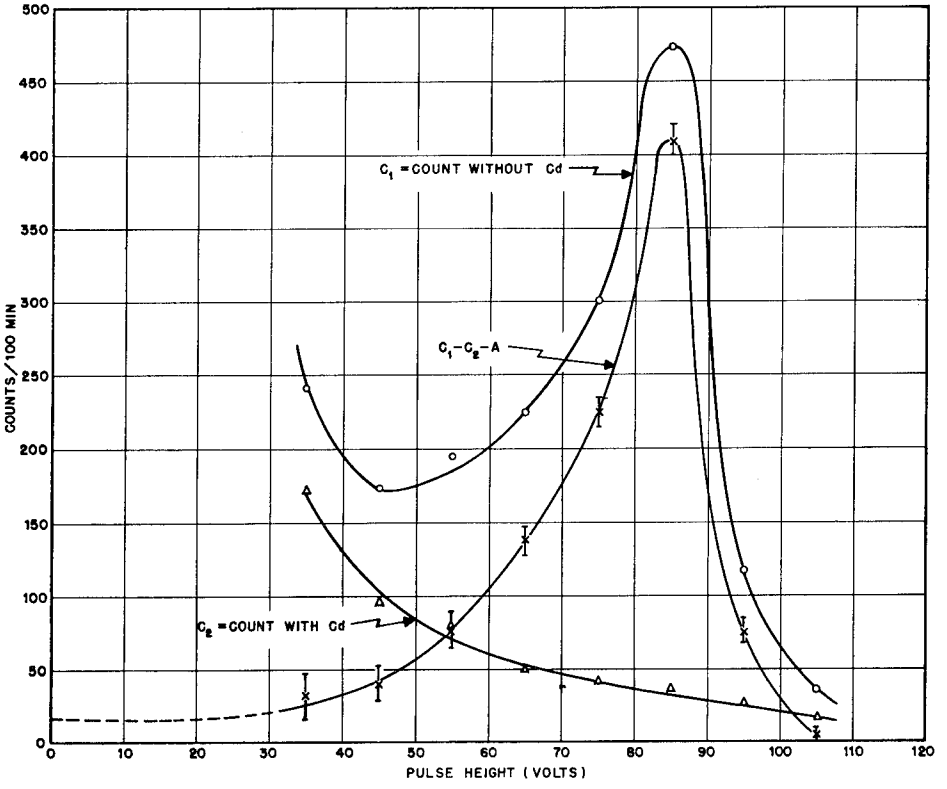

Figure 4. Pulse height distribution curves obtained with "atmospheric" helium. The plotted points represent the number of pulses in a 10 volt pulse height interval and the point is plotted at the center of the interval. The difference curve, $C_1 - C_2 - A$, corresponds to the number of $He^3(n,p)H^3$ disintegrations.

pure argon in the counter. This count with argon serves as a background to be subtracted from the cadmium difference curves gotten with helium.

It is seen that the pulse height distribution curves are not the sharp peaks which one might hope to get with a discrete proton group. The peaks suffer a spread typical of proportional counters.

An approximate "wall effect" correction was calculated for each gas filling taking account of the range of the proton and the stopping power of the gas. In this calculation it was assumed that the angular distribution of the protons was isotropic, and the following approximations were made: the counter walls were considered as plane instead of curved, the effect of corners was neglected, contribution of the heavy recoil nucleus was neglected, the specific ionization of the protons was assumed independent of energy. Let N_0 be the number of disintegrations per cm^3 in the counter, and let R be the path length of a disintegration proton which expends all its energy in the gas. It can be shown that under the above approximations, the number of protons striking the counter wall per cm^2 is $N_0R/4$. The pulse height distribution arising from these protons is such that there are an equal number of pulses per unit pulse height interval for pulse heights ranging from zero to a maximum value, this maximum value being the pulse height arising from a proton which loses all its energy in the gas. By use of this calculation the measured pulse height distribution curves were extrapolated to zero pulse height as indicated in Figures 2, 3, and 4.

The number of disintegrations D taking place within the sensitive volume of the counter was obtained from:

$$D = \Sigma C_1 - \Sigma C_2 - \Sigma A + w$$

where ΣC_1 is the counting rate without cadmium summed over all channels counted, ΣC_2 is the counting rate with cadmium summed over all channels counted, A is the background as determined with pure argon, and w is the "wall effect" correction determined by adding up the number of pulses under the extrapolated lower end of the corrected pulse height distribution curves. The correction w accounts at least in part for all small pulses whether they are due to "wall effect" or to other causes which reduce their size so much that they cannot be counted in the lowest pulse height counting channel.

Inserting values for p and D from Table 1 into relation (2), using $\sigma_{He^3}/\sigma_N = 4300$, and solving for I gives values listed in Table 2, along with values obtained by Aldrich and Nier[5] who have analyzed various mineral sources of helium by mass spectrograph and find a wide fluctuation in He^3 abundance. For correlating the He^3 abundance with the source of helium, the mass spectrograph is better than the present counter technique because of the large quantity of gas necessary in the counter method. For analysis of the He^3 content in enriched samples the counter technique may offer some advantages in ease and accuracy, depending of course on available facilities.

The accuracy of the present measurement is estimated at $\pm 15\%$, which does not include the error in the value of σ_{He^3}/σ_N given as $\pm 20\%$. Of the 15% error, approximately 6 % is statistical error determined by considering the number of counts obtained. The correction w which includes "wall effect" may introduce 5% error. Background fluctuations from spurious electrical disturbances assume more importance because of the low counting rates. There is an undetermined effect of capture gammas from neutron capture in cadmium; these gammas cause a larger number of small pulses when cadmium surrounds the counter than when it does not.

Thanks are due to Richard Taschek and E. R. Graves for helpful criticisms, and to R. W. Davis, Warren Nyer, and Ralph Nobles for help in developing and assembling the apparatus.

Table 1.

	Partial press. p of gas in atmospheres	Press. of added argon	Counter voltage	Figure on which data are shown	Additive correction w	Corrected counts per min = D
N_2 - Run 1	0.258	1.71	3,725	--	137	1314 } Av =
Run 2	0.258	1.71	3,725	2	111	1243 } 1278
"Well" He	7.47	0.27	2,010	3	0.32	9.15
"Atmospheric" He	1.143	0.65	1,485	4	0.54	10.55
Pure A	1.95	--	2,670	3	--	--

Table 2.

	Present measurement	Aldrich and Nier
"Well" helium (Amarillo, Texas)	1.15×10^{-7}	1.4×10^{-7}
"Atmospheric" helium (Airco)	8.6×10^{-7}	12×10^{-7}
Beryl crystals (Amarillo, Texas)		0.6, 1.4, 1.4, 2.0×10^{-5}
Radioactive ores		0.3×10^{-7}

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