INVESTIGATION ON THE ENERGY DISTRIBUTION OF NEUTRONS FROM DISINTEGRATION OF NITROGEN, BERYLLIUM AND FLUORINE

Thesis by

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I N D E X

INTRODUCTION

Artificial transmutation of elements was first observed by Rutherford[1] while bombarding nitrogen with high energy α particles. Since a heavy particle was emitted with a greater range than the bombarding α particle and with even greater range than a recoil proton knocked forward by the head-on collision of an α particle, it was inferred that the α particle had been captured by $^{14}_{7}N$ and a proton emitted.

Soon afterward this investigation was extended to other elements of low atomic number by different investigators. Bothe and Becker[2] worked on the effects of bombarding various elements with α particles from polonium which has the advantage of being free from β or γ ray emission. They found that Li, F, B, and particularly Be gave off very penetrating radiation which they assumed to be γ rays. Mme. and M. Joliot[3] used a strong source of Po and showed that the secondary radiation from Be could penetrate several centimeters of lead and could eject protons from substances containing hydrogen. They measured the energy of the ejected protons in a cloud chamber and suggested that its ejection was a sort of Compton effect. On this basis they estimated an energy of 50 MV for the radiation from Be.

Chadwick[4] observed that the radiation transferred energy to other nuclei also and its energy, calculated on the assumption of conservation of energy and momentum between the photon and nucleus, was different for different recoil nuclei. From the known masses of ^{4}He, ^{9}Be and ^{13}C he concluded that the energy of the radiation could not be greater than 14 MV. He suggested that all the difficulty disappeared if one assumed that a particle was emitted from Be, whose mass was approximately one on the atomic weight scale, to satisfy the conservation laws, and whose charge was zero, to explain the great penetrating power. Since then the masses of nuclei have been measured more accurately and the mass of the neutron has been calculated from the known masses and energies of other particles involved in disintegrations assuming the conservation of energy and momentum. The mass actually adopted is 1.0090. The distribution of neutron energies from some light elements bombarded with natural radioactive particles and the efficiency of disintegration by bombarding particles of different energies has been investigated by Chadwick, the Joliots, Dunning, Bonner, and Mott-Smith.

The height of the potential barrier of nuclei is at least several Mv. According to older views, the bombarding particles should have an energy more than this in order to penetrate the potential barrier. But according to the principles of wave mechanics and Gamow's theory, particles with energy far less than that sufficient to penetrate the potential barrier still have a finite chance of penetration. If, therefore, the number of artificially accelerated particles is large enough, one may expect nuclear transformations with energies far below the barrier height.

The first transmutation with a non-radioactive source was carried out by Cockroft and Walton(5). In their apparatus, the potential generated by a transformer was rectified and multiplied several times by an arrangement of valves and condensers. By bombarding Li by H they observed the emission of α particles of 8.4 cm. range.

Following Cockroft and Walton, Lawrence and Livingston(6) with the aid of a cyclotron accelerated newly discovered heavy hydrogen and obtained evidence for the disintegration of many more elements. They have accelerated deuterons up to 6 MV.

Lauritsen and his associates(7) constructed a million-volt tube and were the first to produce transmutations yielding neutrons by using artificially accelerated particles. They investigated the efficiency of bombarding particles with different energies and the yield of Be bombarded with helium ions and Be, B, and Li bombarded with deuterons.

Later Bonner and Brubaker(8) with the aid of the Lauritsen apparatus and a high pressure automatic cloud chamber, determined the energy distributions of the neutrons from the disintegrations of deuterium, beryllium, lithium, boron, and carbon bombarded with deuterons.

The artificial disintegrations produced by artificially accelerated ions extended the earlier results obtained with α particles from radioactive elements and made possible a whole series of new transformations. The known energy of bombarding particles and the measured energies of those that result from the reaction give relations between the masses of the different particles involved, through the relationship between mass and energy,

$$E = mc^2$$

The plot of the number of yielded particles against their energy in general shows the existence of groups, which indicates that after a particle has been ejected the nucleus is sometimes left in an excited state. This excited nucleus either goes to a more stable state with the emission of a γ ray or further disintegrates. The investigation of the effectiveness of bombarding particles of different energies in disintegration gives information about resonance levels and the height of the potential barrier of the bombarded nucleus.

The above types of information, along with others, have thrown light on the mechanics of disintegrations, interaction between different particles, degree of stability of nuclei, and their possible energy states.

The transformations that we were investigating were mostly those in which light elements were bombarded by particles artificially accelerated in a discharge tube and neutrons ejected. The bombarding particles that we used were deuterons and helium ions, accelerated to a peak voltage of about one million volts.

In general, the higher the atomic number of an element is, the greater is the voltage required for its disintegration. So we expect smaller yields for the same voltage, in higher elements, and in these cases the effect of contamination on the target or in the ion beam is very important as it might mask completely the result under consideration

APPARATUS FOR ACCELERATING IONS

The tubes and ion sources used in our experiments were: one constructed by Lauritsen and his associates,[1] and another short ion path tube built by Stephens, both set in the High Voltage Laboratory.

They were pumped out to a pressure of 10^{-5} mm. of Hg. by a Megavac forepump and three metal oil diffusion pumps, a 7", 3" and $1\frac{1}{2}$" pump in series. While running, however, the pressure was around $3 \cdot 10^{-4}$ mm. Hg. due to the gas supply to the ion source. These pressures were measured by a McLeod gauge and the variation in pressure was checked with a pirani resistance gauge which read down to 10^{-5} mm. Hg.

The tubes can be run up to a million volt peak voltage, which was supplied by a cascaded set of high voltage transformers. The tubes have multiple electrodes on which part or full potential is applied and the ions are accelerated in several stages by passing the gaps at the ends of the hollow electrodes. The high voltage was measured indirectly with a voltmeter which reads the voltage on an auxilliary winding on the first transformer. This voltmeter was calibrated with a sphere gap spark as primary standard. The target tube was insulated from the accelerating tube and was grounded through a microammeter in order to measure the ion current to the target. A magnet was placed above the target tube to deflect the electrons which came down during one half of the cycle, and thick pieces of lead shielded the cloud chamber and experimenting room from the x-rays which the electrons produced.

The ion source consisted of a filament, copper water-cooled anode, and probe electrode supported by two insulating glass rings. A potential of 400 volts was applied between the filament and the anode and a potential of 1400 volts between anode and probe electrodes. A generator mounted on top of the tube and driven through a long insulating belt from the ground supplied the necessary power for the potentials and filament current. The ion source provided a current of positive ions of about one milliampere. The current hitting the target contained molecular and atomic ions of the gas that was supplied to the ion source and ions of any contaminating gases such as hydrogen, nitrogen and oxygen. When deuterium was being used we believe that only about four percent of the total ion current consisted of D^+ ions.

DETECTING AND MEASURING THE NEUTRON ENERGIES

Neutrons are observed indirectly as a result of collision with other nuclei. By considering the laws of conservation of energy and momentum, in the case of elastic collisions between neutrons and nuclei, the energy transferred by the former to the latter can be calculated

$$E_i = \frac{4M_i\ M_n\ E_n \cos^2\varphi}{(M_i + M_n)^2}$$

where M_i, E_i and M_n, E_n are the masses and energies of the nucleus and neutron, and φ is the angle between the momentum of the incident neutron and the recoil nucleus.

The automatic high pressure cloud chamber built by Bonner and Brubaker[8], filled with different gases at different pressures up to 16 atmospheres, was used to measure the energies of those recoil nuclei. Three stereoscopic pictures of the tracks were taken with the aid of two mirrors placed on opposite sides of the chamber. A motion picture camera, loaded with 32 mm. panchromatic supersensitive film was used to photograph the chamber which was strongly illuminated with a horizontal beam of light from a 2 KW. movie flood lamp that was flashed to full brightness when the shutter of the camera was opened. The expansion of the chamber, the removal of the sweeping field, the opening of the shutter, etc., were controlled automatically with the aid of a timer and a series of contacts. The vapor to be supersaturated was supplied from the bottom part of the cloud chamber. Recently we have used a mixture of three parts of ethyl alcohol to one part of water since this vapor required a smaller expansion ratio to be condensed, about 1.15. The gas ordinarily used in the chamber was a mixture of 84.8% methane, 14.4% ethane, and 0.8% nitrogen. The stopping power of this mixture was computed from the pressure read on a calibrated pressure gauge and from data given by Bragg:

$$0.86 \text{ for } CH_4, \quad 1.52 \text{ for } C_2H_6, \quad 0.99 \text{ for } N_2.$$

For ethyl alcohol vapor, a stopping power of 1.92 was calculated from Phillip's[11] data. These stopping powers are compared to air and are for 15°C. and 760 mm. Hg. The energy range data and the stopping powers of mica and helium were taken from Mano's paper [12].

EXPERIMENTAL PROCEDURE

When the vacuum condition of the tube was satisfactory, the electrodes were first freed from adsorbed gases by running the tube with increasing voltage. Then the gas fromwhich the ions were to be made was fed into the ion source through an adjustable needle valve. The filament current and probe voltage were on continuously while the tube was running, but the plate voltage was flashed on for about .05 second so that positive ions hit the target only when the chamber was fully expanded and still sensitive. Then 0.1 seconds later, when the tracks were broad enough but not yet distorted, the camera shutter was opened and stereoscopic pictures of the tracks were taken. The images of the tracks were reconstructed later by the same frame and mirrors that were used in taking the pictures and the true lengths of the tracks were measured.

THE ENERGY DISTRIBUTION OF NEUTRONS FROM NITROGEN BOMBARDED WITH DEUTERONS

The nuclear reaction, with emission of neutrons, that takes place when nitrogen is bombarded with deuterons is:

$$^{14}_{7}N + ^{2}_{1}H \rightarrow ^{15}_{8}O + ^{1}_{0}n$$

The $^{15}_{8}O$ later goes to $^{15}_{7}N$ with the emission of a positron.

The target used most of the time was $NaNO_2$. Ammonium salts were not durable and the yield fell to zero rapidly as the salts sublimed where the ion beam hit.

NaN_3 was very unstable; examination after a short time showed that the salt was completely decomposed and a layer of sodium remained on top of the target.

The ions were accelerated up to a peak voltage of 930 KV and the target current was several microamperes of unresolved ions.

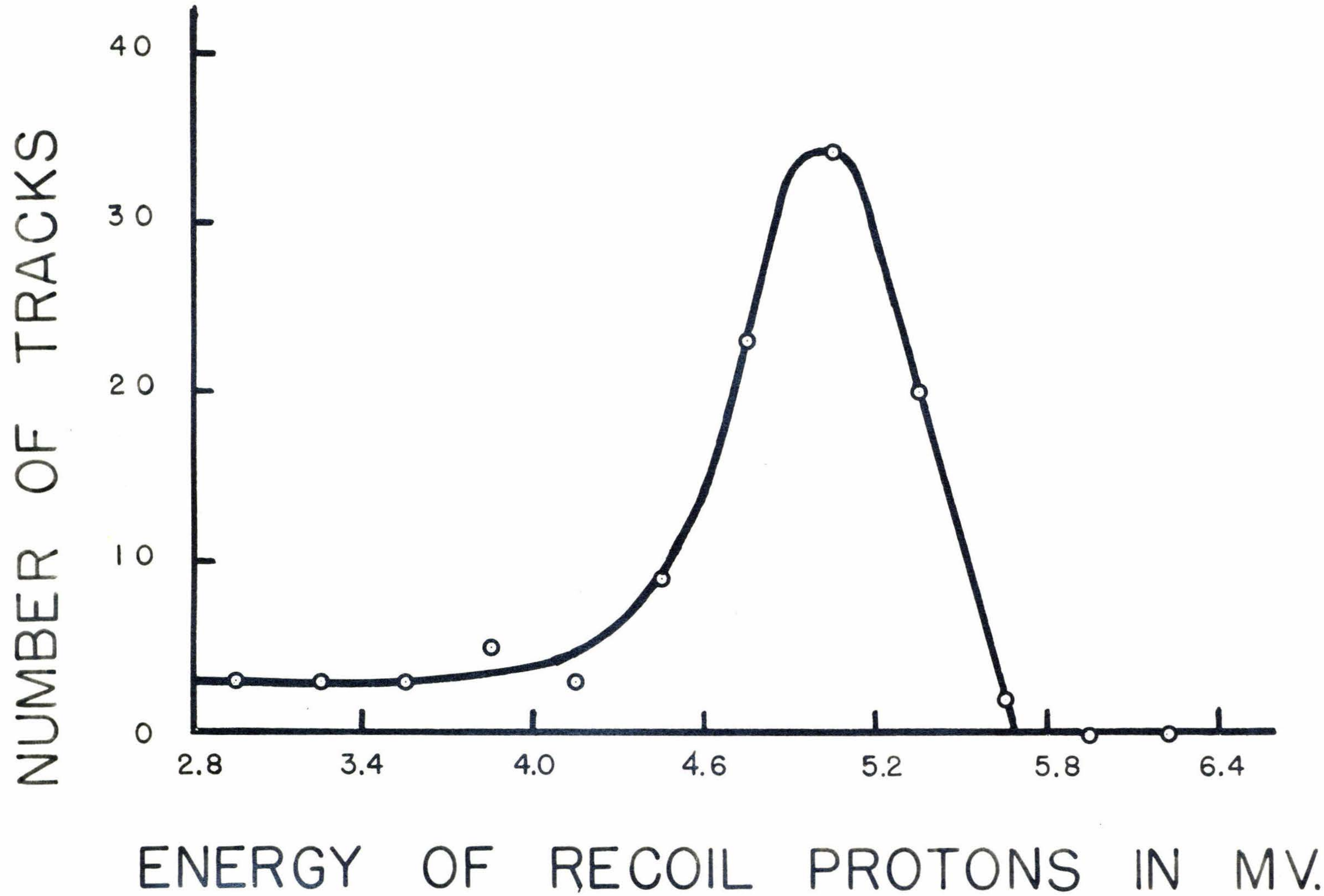
Three series of runs were made:

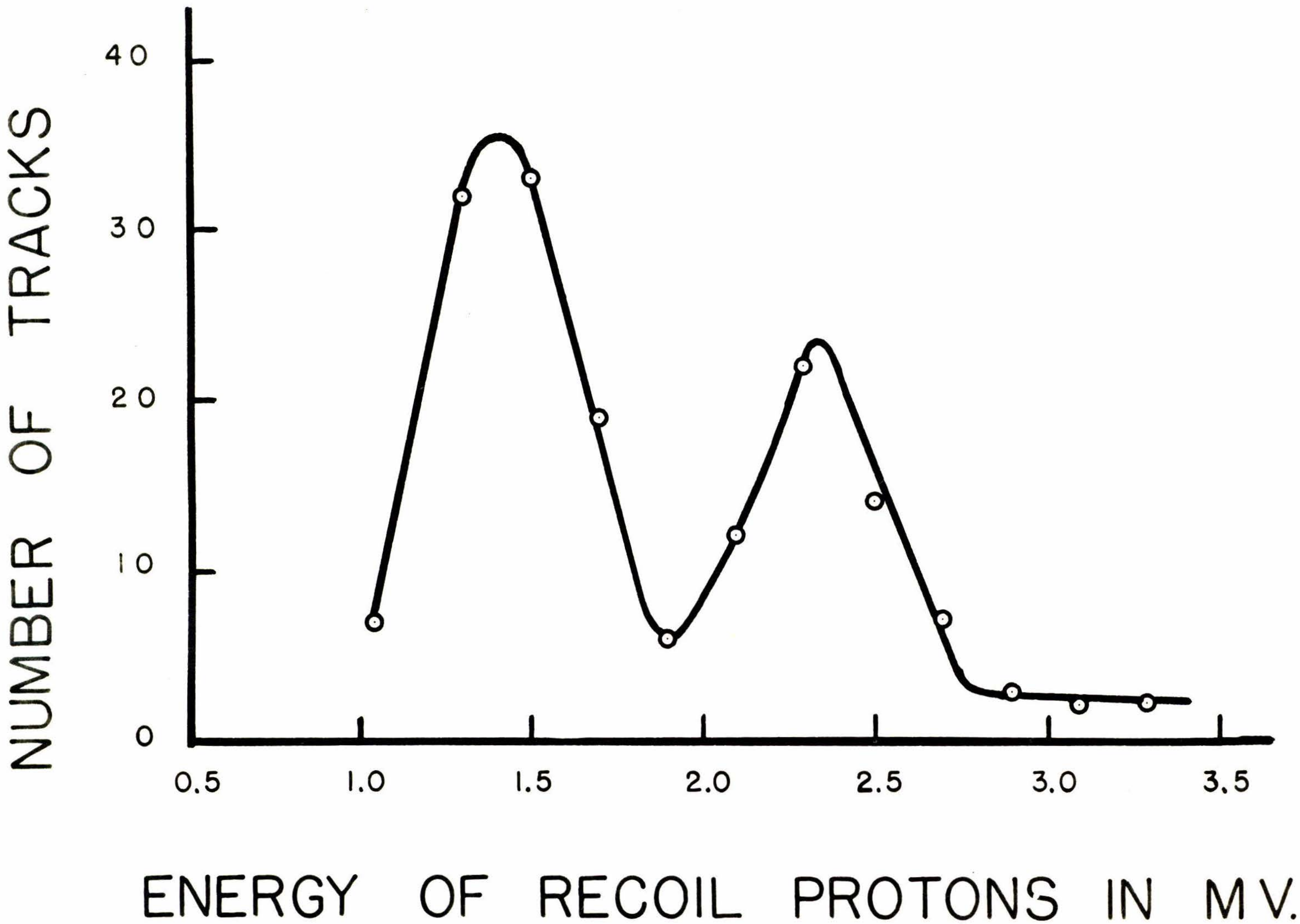
1 - In order to have more accurate data concerning the low energy region, the cloud chamber was filled with hydrogen at 11.4 atmospheres, and 7500 sets of stereoscopic pictures were taken. On these pictures 80 tracks were measured. The stopping power of the gas was calculated from the range of α particles from a polonium source.

2 - The cloud chamber was filled with illuminating gas at 10.5 atmospheres and 6000 pictures were taken, on which 164 tracks were measured.

The results of above measurements with some of the previous work on this problem are shown in Figures I and II.

These curves represent the number of tracks plotted against their energies, and indicate three distinct groups of energies 1.9, 2.7 and 5.7 MEV with relative abundance 3:1:2.


NUMBER OF TRACKS
40
30
20
10
0
2.8
3.4
4.0
4.6
5.2
5.8
6.4
ENERGY OF RECOIL PROTONS IN MV.


NUMBER OF TRACKS
40
30
20
10
0
0.5
1.0
1.5
2.0
2.5
3.0
3.5
ENERGY OF RECOIL PROTONS IN MV.

3 - the 2.7 MEV group has the same energy as $D+H^1$ reaction. A series of runs at 720 peak voltage showed an increase by a factor 10 in relative abundance of the 2.7 and 5.7 MEV groups.

Assuming the excitation curve for nitrogen at this energy region being very steep compared to the deuterium one, there would be but little doubt that the 2.7 MEV group is from the deuterium reaction.

The yield of high energy neutrons from an Atcheson graphite target under similar conditions is much smaller than from a $NaNO^2$ target so the possibility of carbon contamination seems to be excluded.

BERYLLIUM BOMBARDED WITH HELIUM IONS

The disintegration of beryllium by α particles from radioactive elements was the experimental basis for the discovery of the neutron.(4) The reaction which yields neutrons is

$$ {}^{9}_{4}Be + {}^{4}_{2}He \rightarrow {}^{12}_{6}C + {}^{1}_{0}n $$

This reaction has been investigated by several authors. In particular, Chadwick,(15) Rasetti,(16) Joliot,(17) Bernardini,(18) and Kirsch(19) used α particles of varying residual ranges and measured the excitation curve of the neutrons. According to Chadwick(15) there is at least one resonance level for α particles of range 1.4 cm, but Bernardini(18) gives the lowest resonance at 2 cm range. In these experiments the yield for less than one cm range α particles was negligible.

Since the available number of α particles from radioactive sources is small compared to ion tubes, Lauritsen(9) and his associates used artificially accelerated He ions to investigate the efficiency of the reaction for ion energies between 600 and 1000 KV. They used an electroscope lined with paraffin to detect and measure the neutrons. According to their data, He ions of the above energies are 1000 times less efficient in disintegrating than deuterons of the same energy. The problem was then undertaken by the author to find the energy distribution curve of these neutrons using a cloud chamber.

With a peak voltage of 900 KV. and several microamperes target current, 13,000 sets of stereoscopic pictures were taken. Of the tracks on these pictures, 212 were measured.

Since the yield is very small, the contamination due to deuterons in the beam should be considered. The distribution curve shows at least three neutron groups below 4.6 MV, corresponding to the deuteron beryllium neutron groups as reported by Bonner and Brubaker. Of all the tracks measured, 11 had energies more than 4.6 MV, which is the maximum energy of neutrons from the deuteron on beryllium neutrons. The longest range proton had an energy of 6.1 MV and the expected maximum energy of neutrons observed at 90^o from beryllium bombarded with He^{++} ions of 1.8 MV as calculated from Bonner's masses is 6.2 MV.

Deuterium had been used in the tube before we started this problem and our data shows a continuous decrease in the number of tracks per film. We then continued this investigation in a newly built tube. With a total target current of 30 microamperes and a peak voltage of 940 KV, the yield was still smaller, one proton track in the chamber for every 300 expansions. So it seems that of the tracks measured less than one-sixth are from the α particle reaction. Most of these come from He^{++} ions which are much more effective than He^{+}, although smaller in number. Further investigation of this problem would require a much larger number of He ions in the beam.

FLUORINE BOMBARDED WITH DEUTERONS

The reaction which yields neutrons is

$$^{19}_{9}F + ^{2}_{1}H \rightarrow ^{20}_{10}Ne + ^{1}_{0}n$$

From the masses of ^{19}F and ^{20}Ne given by Aston[20] and Pollard[21], the energy released in the reaction is 10.8 MV and the maximum energy of the neutron emitted at right angles to the bombarding particles direction is 11.1 MV.

We made several series of runs with different pressure and gases in the cloud chamber. A calcium fluoride target was bombarded with deuterons accelerated to a peak voltage up to 970 KV and total target current of more than 40 μa. In order to measure the low energy neutrons more accurately, the chamber was filled with the mixture of methane that we ordinarily use at 6 atmospheres

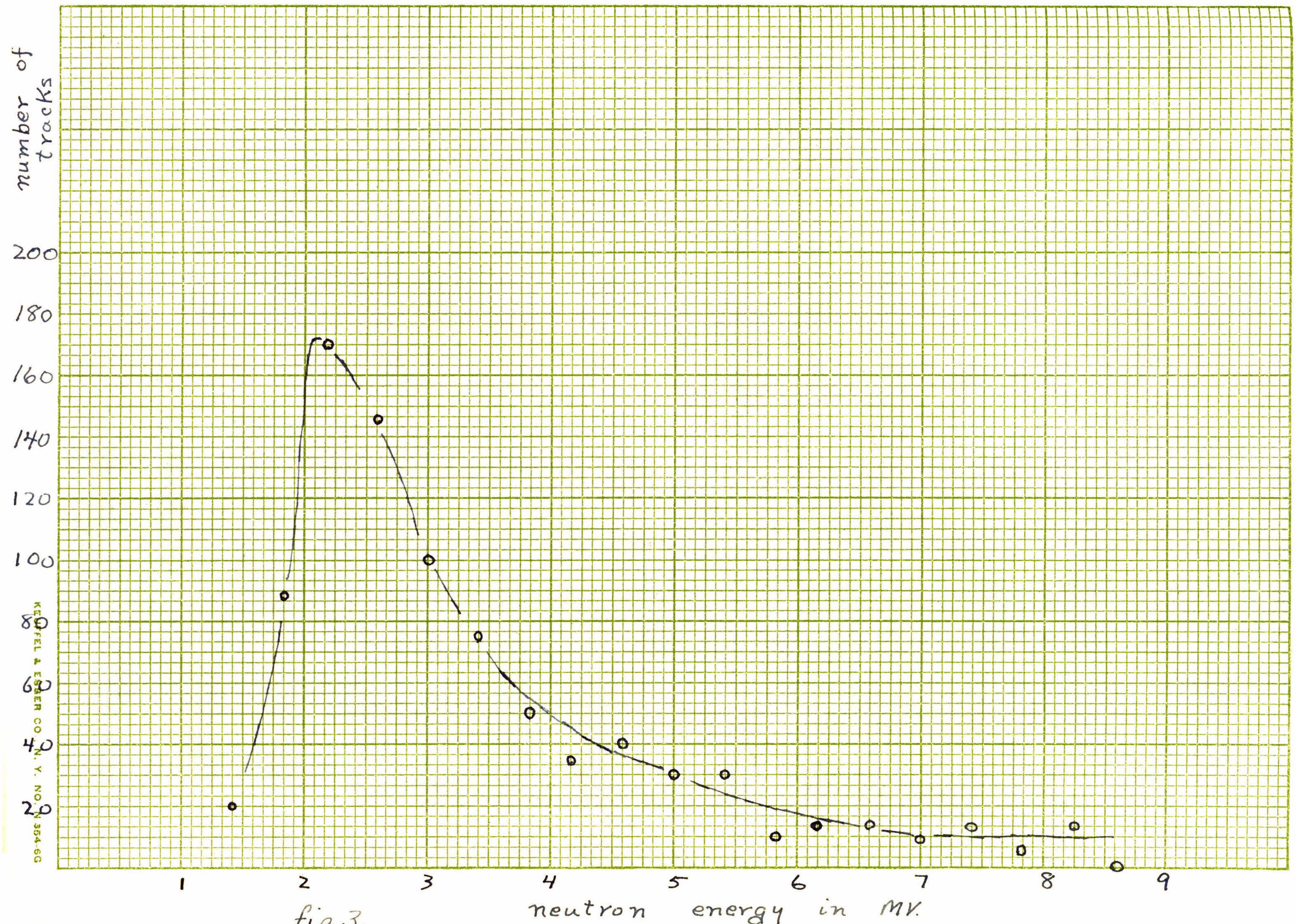

number of tracks
200
180
160
140
120
100
80
60
40
20
1
2
3
4
5
6
7
8
9
neutron energy in MV.
KEUFFEL & ESSER CO. N. Y. NO. N 354-6G

fig. 3.

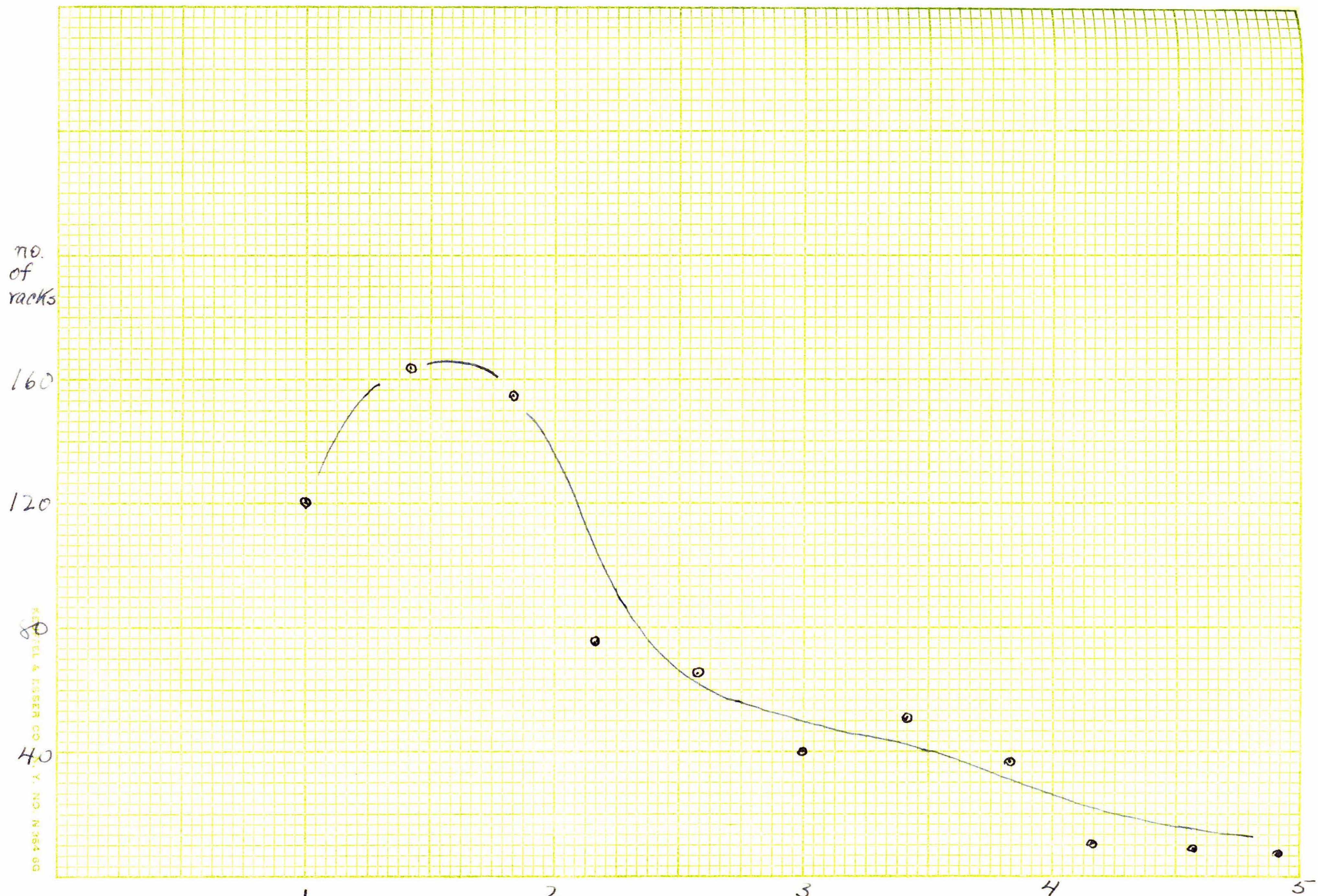

no. of racks
160
120
80
40
1
2
3
4
5
neutron energy in MV
KEUFFEL & ESSER CO. N. Y. NO. N 364 6G

fig. 4

pressure which had an equivalent stopping power of 5.7.

From 3250 sets of stereoscopic pictures we measured 243 tracks up to 5 MV energy. The number against energy curve is shown in Figure 4. Then we increased the pressure to an equivalent stopping power of 15.7 and took a series of 2750 pictures, on which 290 tracks were measured. This distribution curve is shown in Figure 3. For measuring the high energy neutrons, we put a mica sheet of 63 cm. stopping power in the chamber which was filled with methane at 16.4 atmospheres pressure. On 2200 pictures we observed only seven tracks which had energies up to 10.5 MV, only one of which was unambiguous. When mica is used in the chamber, several series of runs with different thicknesses of mica and different pressures in the chamber should be made in order to overlap different distributions.

To get directly the whole distribution curve, the chamber was filled with helium at 10.8 atmospheres pressure which gave a total stopping power of 2.04. In head-on collisions with helium nuclei, the neutron transfers 16/25 of its energy to the α particle, and α particle ranges are about one-fourth those of protons of the same energy. Since there is alcohol vapor in the chamber, some of the tracks in this case are recoil protons and in order to get the true distribution curve, one has to get relatively large numbers of tracks, thereby masking the background due to protons. In our case, we took only 1750 pictures on which 28 tracks were measured. Since the yield was very small, it seemed more convenient to use methane and mica.

From our data, we cannot definitely say how many groups of neutrons there are, but the curves show that a large proportion of the neutrons have energies less than 3 MV. This may include some from deuterom contamination on the target.

PHOTODISINTEGRATION OF Be BY γ RAYS FROM THORIUM C

Chadwick and Goldhaber[22] were the first to observe a nuclear disintegration produced by γ rays. They studied the disintegration of deuterium and beryllium. An ionization chamber connected to a linear valve amplifier and oscillograph was used to detect the charged particles from the disintegration. For detection of the photoneutrons, they used an ionization chamber coated with a layer of lithium metal or boron powder. In the case of Be disintegration they made use of the induced activity in silver to detect the neutrons.

Szilard and Chalmers[23], who were the first to show the disintegration of Be by γ rays from radium C, observed the photoneutrons by means of the induced activity in iodine.

According to Chadwick and Goldhaber, of the two possible processes

$$^{9}_{4}Be + hv \rightarrow {}^{8}_{4}Be + {}^{1}_{0}n$$

$$^{9}_{4}Be + hv \rightarrow 2\,{}^{4}_{2}He + {}^{1}_{0}n$$

the second is predominant, and so one expects a continuous distribution in energy of the neutrons. The most energetic γ rays available from radioactive sources are those from Th C^{11} which have an energy of 2.62 MV. The γ rays from artificial disintegrations have much higher energies, but they are usually accompanied by neutrons from the same reaction or contamination reactions. Since the cross section for photodisintegration is very small, the expected effect from photoneutrons may be less than that obtained from the neutrons accompanying the artificially produced γ rays.

We used 20 mc. of radiothorium in equilibrium with its decay products as a source of γ rays. It was surrounded by 300 gms. of beryllium and placed 7 inches from the cloud chamber. To reduce the γ rays in the chamber about two inches of lead were interposed between the source and the chamber. The chamber was filled with hydrogen at an equivalent stopping power of 1.15. On the 2000 pictures taken, we observed 32 tracks. Of these a few had ranges below the expected maximum of .66 cm and were in a direction from the source. They were more or less continuously distributed as would be expected from neutron scattering in the lead. Some were probably α particles from polonium contamination in the chamber since on 450 pictures taken without the radiothorium we found five tracks.

SUMMARY

The historical development of nuclear transmutations with artificially accelerated ions is outlined. The apparatus which we used for producing disintegrations and measuring the neutron energies is described.

The energy distribution of the neutrons from the disintegration of N by 0.9_3MV deuterons as determined by the method of recoil protons in a cloud chamber, is shown. The energy distribution curves indicate two neutron groups with energies of 1.9 and 5.7 MEV.

The energy distribution curves of neutrons from Be bombarded with helium ions shows three groups of neutrons from beryllium bombarded with the deuteron contamination in the beam, and several neutrons with higher energies up to 6.1 MV. Some of these are due to doubly ionized helium ions in the beam. The results of the experiment indicate that the yield of this reaction is less than 1/1000 of the deuteron beryllium reaction. The contamination of this last reaction masks the first one unless special care is taken.

Fluorine was bombarded with deuterons and the energy distribution of the neutrons was obtained. It does not indicate definitely how many groups there are, but shows that the greater number of neutrons have energies smaller than 3 MV.

Photoneutrons from the disintegration of beryllium by γ rays from radiothorium was looked for in the cloud chamber. Many of the tracks in the chamber were probably due to polonium contamination. All the tracks were distributed more or less continuously.

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