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ABSTRACT

A neutron spectrometer has been constructed consisting of a phototube viewing a plastic scintillator that is surrounded by a BF_3 proportional counter. When a neutron produces a recoil in the plastic scintillator and this neutron is subsequently captured in the BF_3 counter, a gate is formed by this delayed coincidence which allows the height of the first plastic scintillator pulse to be analyzed by a pulse-height analyzer. During this delay time the first pulse has been stored in a delay line while the associated neutron has become thermalized and captured. The spectrometer is essentially nondirectional, and from experimental evidence its lower limit of response appears to be ~3 Mev, with the upper limit (not yet investigated) probably extending as high as 15 Mev. Spectra of Po-Be and Mock Fission sources are included. Monoenergetic neutrons from $d(d,n)He^3$ and $T(d,n)He^4$ reactions were used to calibrate the counter. An interpretation of these results is given.

PREFACE

To investigate the properties of a neutron flux field it would be highly desirable to have a neutron detector whose response gives some information about the energy of the incident neutrons. This paper describes a double-pulse total-absorption type of neutron spectrometer which, it is hoped, will be useful for surveys of neutron fields containing neutrons whose energies lie between 1 and 30 Mev. In addition, this spectrometer has the property of being nondirectional, and thus need not be aligned with the source of neutrons in any specified manner. This is a particularly useful property when either the location of the source is unknown, the source is so large and unsymmetrical as to make collimation impractical, or the neutron flux is falling on the spectrometer from several directions simultaneously.

The author wishes to express his thanks and appreciation to Professor Burton J. Moyer for his encouragement and supervision, and to Dr. Roger W. Wallace for his continual guidance and criticism. In particular, the author thanks Alan Smith, Physicist, who was consulted constantly for background information, and who worked in conjunction with the author on the experimental phases of the project. The author also thanks Dr. E. Milne for his review and final criticism of this paper. To the crews of the Linear Accelerator at Berkeley, and the Cockcroft-Walton Accelerator at UCRL, Livermore, the author expresses his appreciation for their aid in the experimental bombardments.

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CHAPTER I

INTRODUCTION

In the energy range of 1 to 30 Mev, successful neutron spectroscopy has hitherto been accomplished only through the use of nuclear emulsions, which require tedious methods of analysis. In addition, the location of the neutron source must be known so that the proton recoil angle in the emulsion may be established, or a very large number of tracks must be scanned. The original purpose of this experiment was to devise a neutron spectrometer useful in the 1-to-30-Mev range which would apply electronic methods of analysis, and be totally independent of source location.

The major features of several proposed neutron spectrometers, from which the ideas embodied in the neutron spectrometer reported here were evolved, are described in this chapter. The detection of alpha particles by scintillation phosphors dates back to Rutherford, but from his time until the development of the modern photomultiplier tube the light flashes had to be counted by eye, and as a result such experimental work was so slow and tedious that this method of particle detection was forsaken in favor of Geiger and proportional counters. With the advent of the phototube it was soon found that individual light flashes could be detected with high efficiency, and many workers--including in particular Kallman[1]--showed that naphthalene and anthracene were transparent to their own radiation and could be used to detect beta and gamma rays. Moreover, the phototube has a great advantage over the visual scintillation method in that it allows fast time coincidences to be made between counters.

Many neutron detectors, including the spectrometer described here, depend on the (n, p) collision process in a hydrogenous material for their action. The (n, p) collision, occurring between two particles of almost equal mass, gives an energy distribution for the recoil protons that is flat from zero up to the energy of the incident neutron. As a

result the pulse-height distribution of a neutron detector that measures the energy of the first-collision recoil protons from an (n, p) process is a rectangle extending from zero up to the incident neutron energy for monoenergetic incident neutrons. If the incident-neutron spectrum is not monoenergetic the pulse-height distribution from the proton-recoil energy detector is the sum of many such rectangles. The neutron spectrum can be secured from such a first proton recoil-energy pulse-height distribution by differentiation. The first recoil is the only one that allows this simple relation; production of more than one recoil proton in the hydrogenous material by one incident neutron complicates the situation.

In the elastic (n, p) collision the energy of the recoil proton is given by $E = E_0 \cos^2 \theta$, where E_0 is the incident neutron energy and θ is the angle between its path and that of the recoil proton. If E is measured by some detector, as is the case in many neutron spectrometers, E_0 can be calculated if θ is known. Thus for measurement of the energy of a neutron two measurements must be made. If the location of the source of neutrons is known, then θ is usually readily available and only E must be measured. If the source location is not known, or--as is often the case--is sufficiently large so that it cannot be considered to be a point, then θ also must be measured by the spectrometer.

The use of hydrogenous scintillation phosphors for detection of fast-neutron recoils is limited by the fact that gamma rays are usually associated with a neutron flux and are detected with about the same efficiency as are the recoil protons.[2] In the neighborhood of 10 Mev, a gamma ray gives a pulse approximately twice as large as that of a proton of equal energy, as seen in Fig. 1. In the absence of gamma rays, however, a single hydrogenous scintillation counter may be used as a spectrometer.[3] Care must be taken in the use of such a spectrometer, because the scintillator response may not be linear; in addition, if the gamma sensitivity can be neglected, sufficient statistics must be collected to allow for differentiation of the original pulse-height distribution. Multiple collisions in the scintillator itself will also make the

analysis difficult, since the resulting proton pulse-height spectrum will no longer be the simple integral of the desired neutron spectrum.

There are several types of neutron spectrometers for the 1-to-20-Mev region which analyze proton recoils from neutrons incident on organic scintillation plastics or crystals.[4] One method utilizes a pulsed source of fast neutrons whose times of flight are measured over a fixed flight path.[5] Draper[6] also discusses an experiment that involves measurement of the energy of recoil of the proton by pulse height in one scintillator while the angle of recoil of the neutron is selected by the requirement of a coincident pulse in a second scintillator. Chagnon, Madansky, and Owen[7] report on a similar spectrometer in which the secondary counter is a ring of organic liquid scintillation counters.

Muehlhause and Thomas[8] discuss briefly a liquid scintillation neutron detector which has an organic compound of boron introduced in a fluorescent hydrogenous medium. When fast neutrons enter this medium, two pulses separated by a time interval calculated to be about 0.5 μsec should be detected. The first pulse is produced by proton recoils and the second pulse is the result of the (n, α) capture of the neutron by the boron present. This double-pulse arrangement discriminates against gamma and slow-neutron background and--in conjunction with time-of-flight equipment--should prove to be an extremely useful technique for neutron spectrum analysis.

Reines et al.[9] describe a similar double-pulse liquid scintillator, which incorporates a cadmium-loaded solution. Ninety type 6292 photomultiplier tubes look at about 50 gallons of this solution. The tubes are operated in two banks in coincidence so as to eliminate tube noise. Once again the first pulse corresponds to the sum of the various recoiling protons and the second pulse to the final neutron capture. The individual proton-recoil pulses arising from the slowing down of the neutron come so close together that they cannot be resolved in time, but they are, as a group, separated from the final capture pulse by several microseconds, since the thermalized neutron wanders around for some time before it is captured by the cadmium.

Smith and Thompson at UCRL, Berkeley experimented with a spectrometer of the double-pulse cadmium-loaded type of liquid scintillator, but used a small volume of liquid viewed by a single 5819 phototube. The small volume was used in order to lower the counting rate and (consequently) the accidental-coincidence rate, which is proportional to the square of the single-counting rate. Simultaneously the resolution of the spectrometer was increased by discrimination against events in which the neutrons incident on the scintillator did not lose a large fraction of their initial energy in the first collision. When a large fraction of the incident energy is transferred to the first recoil proton, there is a greatly increased chance of the remaining neutron's colliding in the scintillator and producing a proton pulse that is too small to be important, but leading to a chain of small recoils which have an increased probability of thermalization and eventual capture of that neutron. Thus large-energy-transfer first-recoil pulses are preferentially selected by a small scintillator volume, when it is used in conjunction with a double-pulse-selecting system that requires eventual thermalization. In principle an optimum volume can be obtained in which the counting rate is high enough to be practical, in which the accidental counting rate is small, and in which multiple scattering is relatively unimportant. If the scintillation pulse-height response were linear with proton energy then multiple collision events in a scintillator large enough to contain all of the recoils from a single neutron (as in the case of the Reines detector) would be acceptable, but in view of the nonlinearity of the response--as seen in Fig. 1--it was considered desirable to discriminate against multiple-recoil events as outlined above.

Difficulties were encountered by Smith and Thompson, since the second of the two pulses was a small pulse in a background of large pulses. The accidental-coincidence counting rate was too high to make this method useful. The pulse size was small because the electron from the gamma released upon cadmium capture was not usually contained in the small volume of liquid.

The $B^{10}(n, \alpha)Li^{7}$ reaction is extensively used in slow-neutron detectors because of its large cross section. The exoergic reaction with Q = 2.78 Mev averages approximately 2.3 Mev of ionization energy, owing to the predominance of the captures leading to the 0.48-Mev excited state of Li^{7}, which returns to the ground state via gamma emission. Up to approximately 1 kev the effective (n, α) cross section of natural boron follows the 1/v law, with σ = 730 barns at thermal neutron velocities. The isotope B^{10} is 18.83% abundant in the natural state, and has an isotopic (n, σ) cross section of σ = 3960 barns. The availability of ~100% B^{10}-enriched boron therefore makes possible the construction of neutron detectors of extremely high efficiency over the lower energy ranges.

Fowler and Tunnicliffe[10] and Rossi and Staub[11] discuss various types of BF_3 proportional counters, including constructional features and precautions to be taken in gas preparation. As survey instruments for the detection of neutrons, BF_3 proportional counters have found no equal; once calibrated with a source of known strength, a counter can be used to determine the slow-neutron flux, and shows negligible sensitivity to gamma rays.

CHAPTER II

THE CONSTRUCTION AND DEVELOPMENT OF THE SPECTROMETER

Following several discussions with workers interested in neutron spectroscopy, the author settled on an idea furnished by Alan Smith[12] for a double-pulse type of spectrometer. Smith had previously done some work with a geometry that included a large plastic scintillator surrounding a small BF_3 counter. It was felt, however, that the overall efficiency of this spectrometer would be extremely small because of the limited solid angle available to a neutron for capture by the BF_3 counter following thermalization in the large scintillator. Also, it is desirable--as discussed previously--that neutrons lose their energy in a minimum of collisions, and having a large scintillator resulted in a poor geometry from this standpoint.

A neutron thermalized in a scintillator has the greatest probability of being captured in a BF_3-filled proportional counter if this counter completely surrounds the scintillator, so I proceeded to design a BF_3 counter that would contain a scintillator and a photomultiplier tube at its approximate center. Spherical designs were discarded as too difficult to manufacture, and a cylindrical arrangement was finally settled upon. Figure 2 shows a cross-section view of the BF_3 counter. As can be seen, the counter consists of an 8-inch-diameter cylindrical tank, with a re-entrant well of 1.6 in. diameter placed along the axis of the 8-inch cylinder, into which the scintillator and phototube may be inserted. The scintillator is a polystyrene cylinder, loaded with terphenyl, tetraphenyl butadiene, and zinc stearate, 2 inches long and 1.5 inches in diameter, which is bonded to a Du Mont 6291 photomultiplier tube by Dow-Corning DC 200 silicone. The scintillator size was chosen with due consideration of the limitations discussed in Chapter I, namely: the smaller the scintillator the higher the probability of a neutron's being moderated by a single collision, and the less the probability that a gamma ray entering the scintillator would be counted; although at the

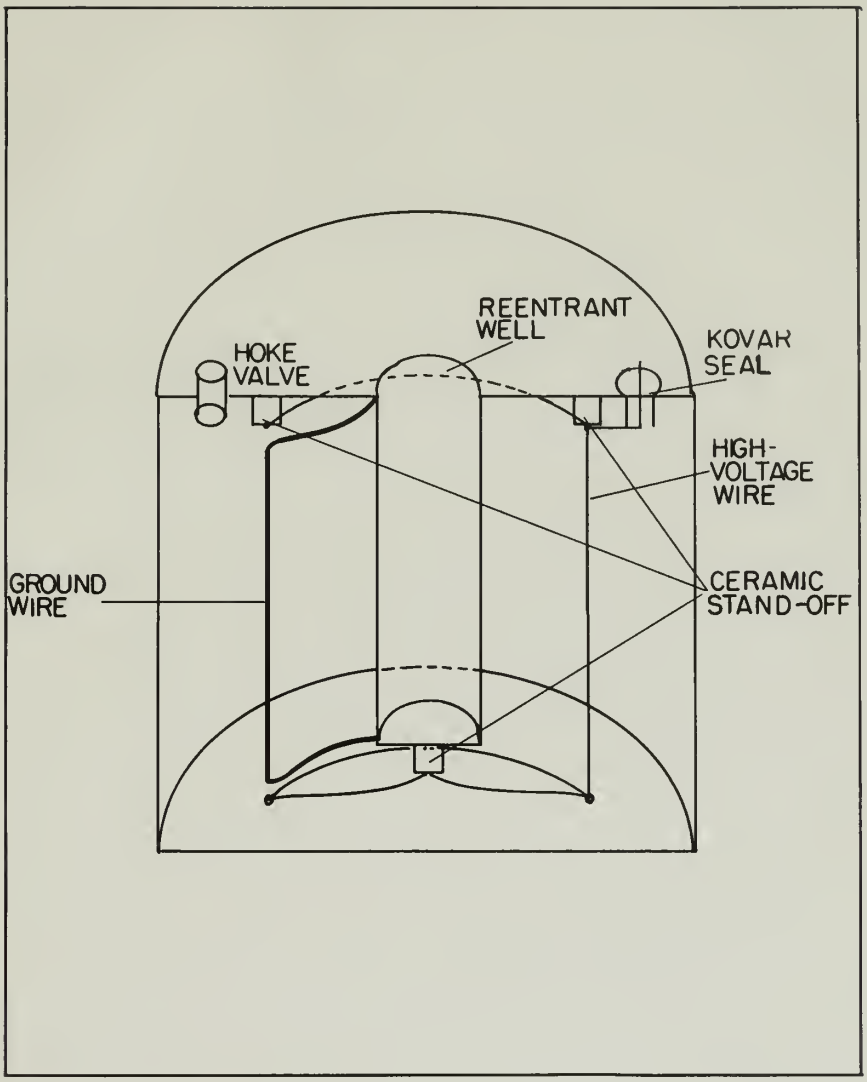

Fig. 2. Cross section of the sensitive BF_3 proportional counter with re-entrant well for insertion of scintillator and 6291 photomultiplier tube. The counter has a total of six high-voltage electrodes evenly spaced at 60° intervals and separated from one another by heavy 0.125" copper ground wires.

same time the small scintillator would be poor with respect to counting rate, and statistics would suffer. The initial scintillator size was therefore a compromise and may have to be adjusted to the counting rate in any particular application. The 6291 tube was chosen mainly because of its small diameter, allowing the over-all outside diameter of the entire counter to be as small as possible. The only surface of the scintillator that is not bounded by BF_3 counter is the face adjacent to the tube, and under any circumstances this would be difficult to cover. With this geometry more than 80% of the thermalized neutrons must eventually migrate into the BF_3 volume.

After several experiments, during which the number of high-voltage wires in the BF_3 counter was changed by a number of stages from one to a total of twelve, a final configuration was decided upon that included six high-voltage wires (3-mil stainless) separated from one another by heavy copper ground wires. This in effect gives a configuration similar to six separate cylindrical counters surrounding a common center axis. During this period the counter was filled with 95% argon and 5% CO_2, and was used in conjunction with a calibrated Po-Be source. With the six hv electrodes in place, and with a filling of argon, the counter was tested for directional properties through the use of a highly collimated source. As desired, the counter proved to be quite nondirectional, being only slightly less sensitive to radiation arriving at the bottom or the top of the cylinder than at the sides.

After the optimum wire configuration was arrived at, the counter was filled to an absolute pressure of 10 cm of Hg with ~100% B^{10}-enriched BF_3 gas. Initial investigations showed that it would be desirable to increase the efficiency of the counter, so the amount of BF_3 gas was increased to 25 cm of Hg (absolute).

The high-voltage and bias characteristics of the counter are shown in Figs. 3 and 4. As can be seen, the range, from about 2150 to 2300 volts, is an excellent high-voltage plateau region. For all future investigations discussed in this paper 2250 volts has been used, with an amplifier gain of about 7000 followed by a discriminator whose bias was

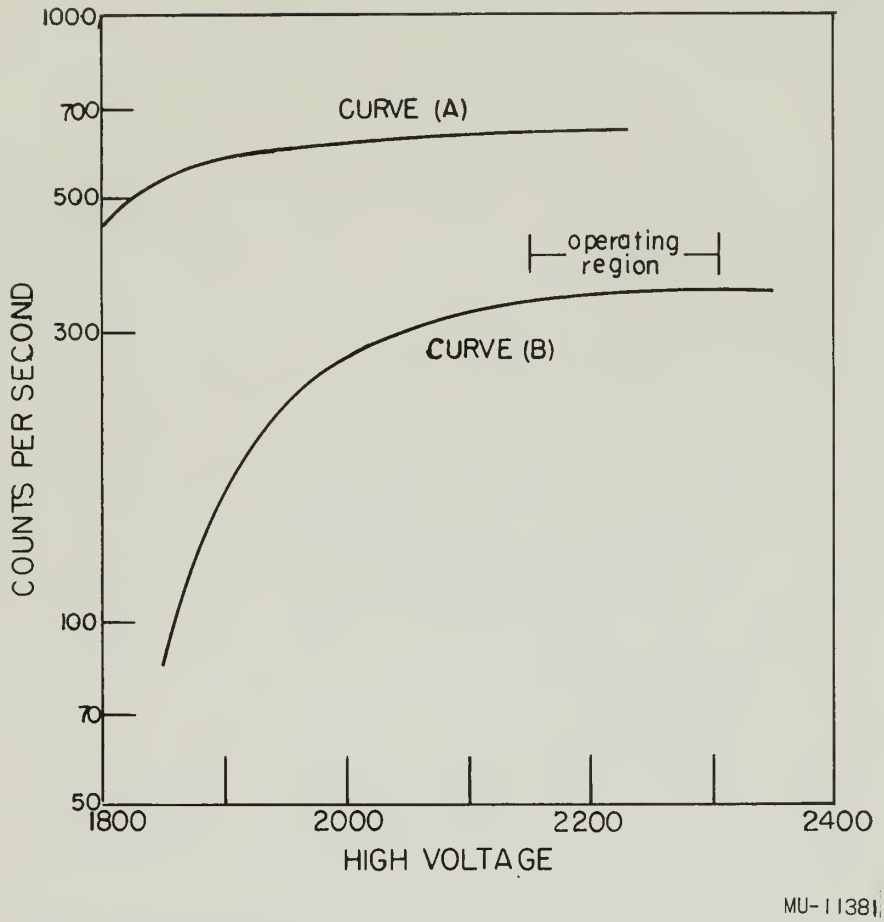

Fig. 3. High-voltage plateau curve for the BF_3 proportional counter with a filling of 25 cm of 100% B^{10}-enriched BF_3 gas, and with an amplifier gain of ~7000. Curve A is the curve of an operating BF_3 survey instrument for slope comparison. Curve B is the curve of the counter used in the system under discussion, and indicates an excellent operating region between 2150 and 2300 volts.

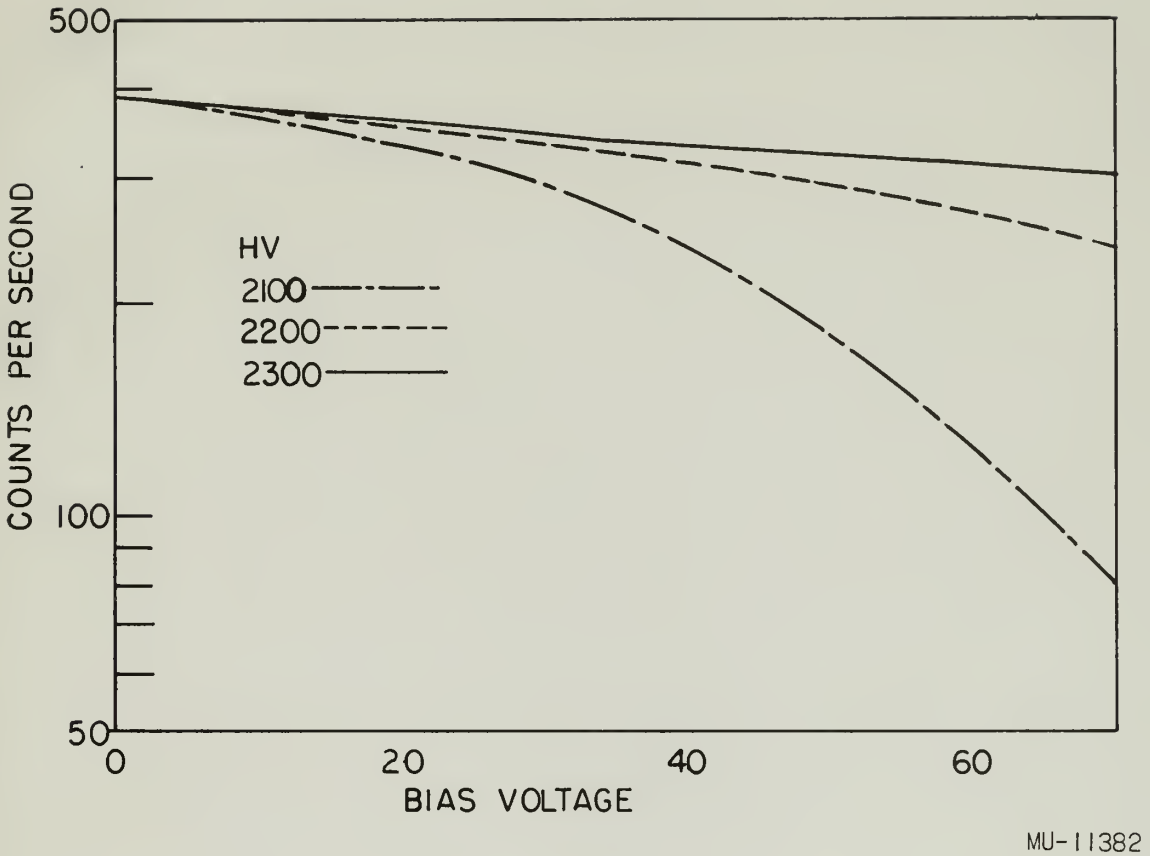

Fig. 4. High voltage-bias voltage curves for BF_3 proportional counter.

set at approximately 20 volts. Under these conditions of operation the counter is gamma-insensitive.

As shown in Chapter I, the photomultiplier pulses were to be delayed before being analyzed in order to allow for the time required for the thermalized neutron to migrate through the crystal and into the BF_3 gas and be captured. For the original crystal (1.5 in. in diameter by 2 in. long) rough calculations gave an average migration time of 30 μsec ($v_\tau = 2.2 \times 10^5$ cm/sec). When a four-channel tandem gate with 5-μsec gate widths was used, triggered by the photomultiplier pulses but counting the BF_3 pulses, the average delay for BF_3 capture was found to be approximately 10 microseconds, as shown in Fig. 5. Owing to difficulties encountered in attempting to retain pulse shape and size and to simultaneously delay these same pulses, the shortest delay practicable should be used.

Figure 6 is a block diagram of the experimental arrangement. When terminated HH-2000 or RG 65 type delay cable was used, it was found that 10 μsec of delay did not alter the pulse height or shape appreciably, and at the same time 50% of the useful BF_3 pulses were counted. This 10-μsec delay for all photomultiplier pulses was thereupon adapted for use in the completed configuration. The BF_3 pulses, after being amplified in a UCRL Model V Linear amplifier, are fed through a standard UCRL 1024 Scaler for counting and pulse shaping. The uniform pulses out of this scaler are fed into a cathode coupled multivibrator for pulse-stretching purposes. This multivibrator puts out an essentially flat pulse of 10 μsec duration, which is fed into a coincidence unit whose other input is the delayed photomultiplier pulse from another UCRL Model V Linear amplifier. The relations of these pulses are shown in Fig. 7. A UCRL scale-of-1000 ten-channel pulse analyzer is gated on by the above coincidence, and the delayed photomultiplier pulse is counted in the appropriate channel according to pulse height. In the work described later, in which the Cockcroft-Walter accelerator at UCRL Livermore was used, a 50-channel pulse-height analyzer was employed. The unshielded counter with all associated electronics was

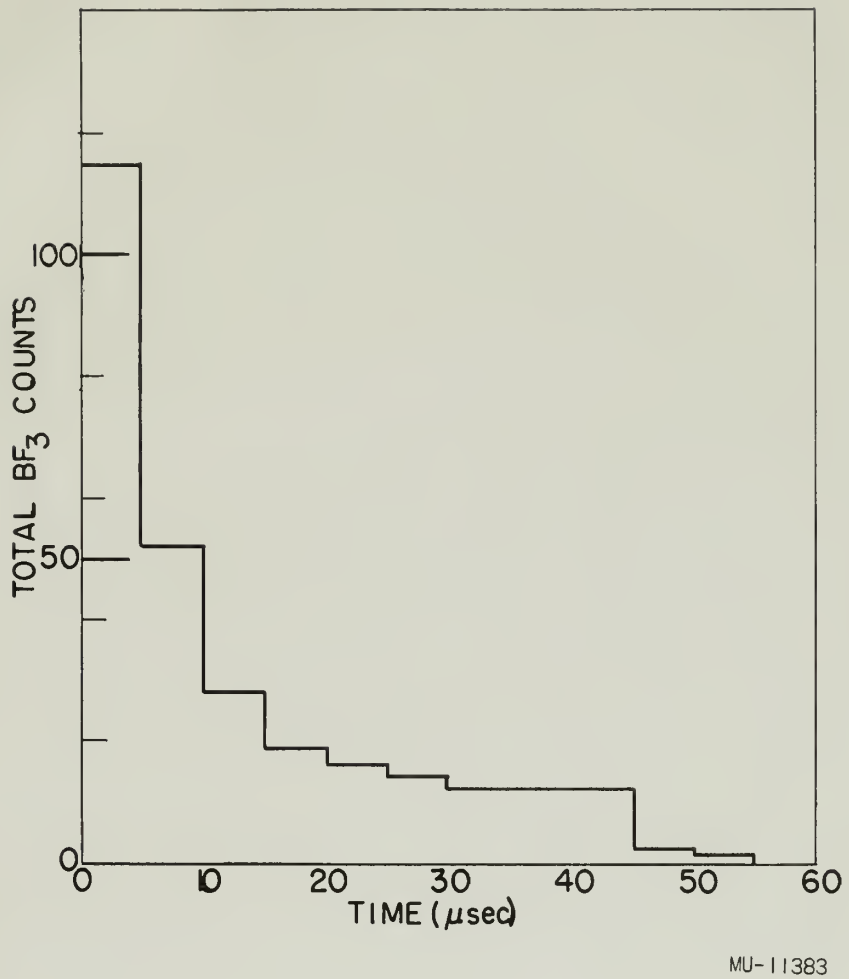

Fig. 5. Delay time for BF_3 capture following thermalization in scintillator. It is noted that ~58% of BF_3 captures occur within 10 μsec.

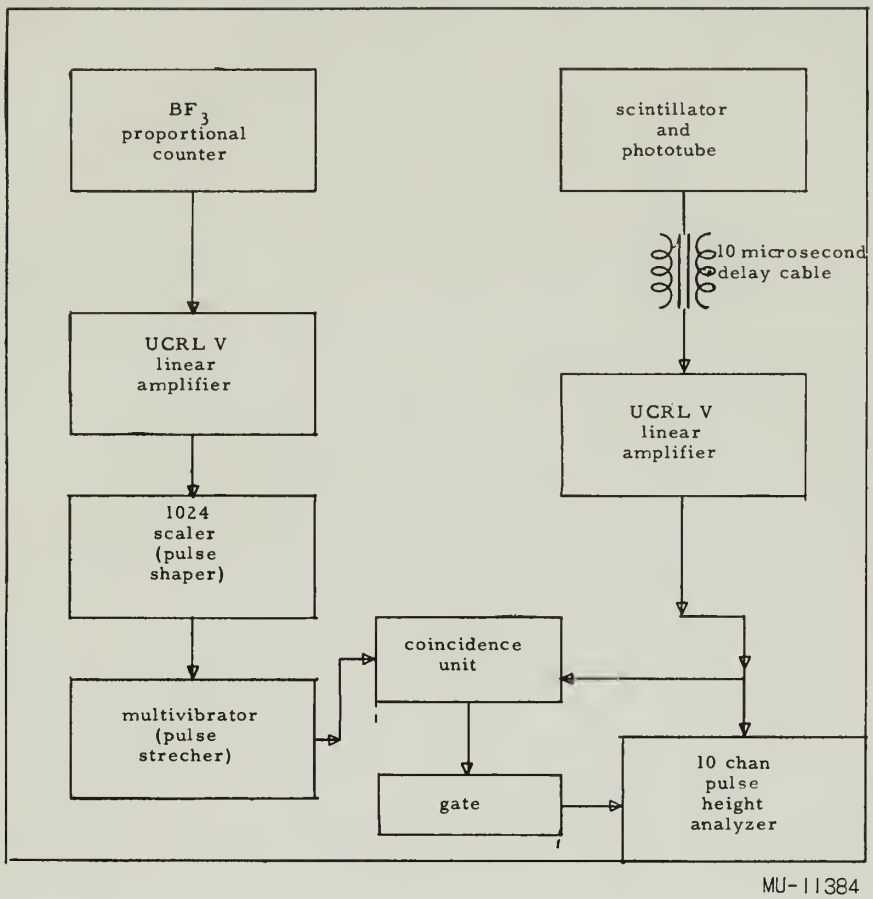

Fig. 6. Block diagram of spectrometer and associated electronic circuitry.

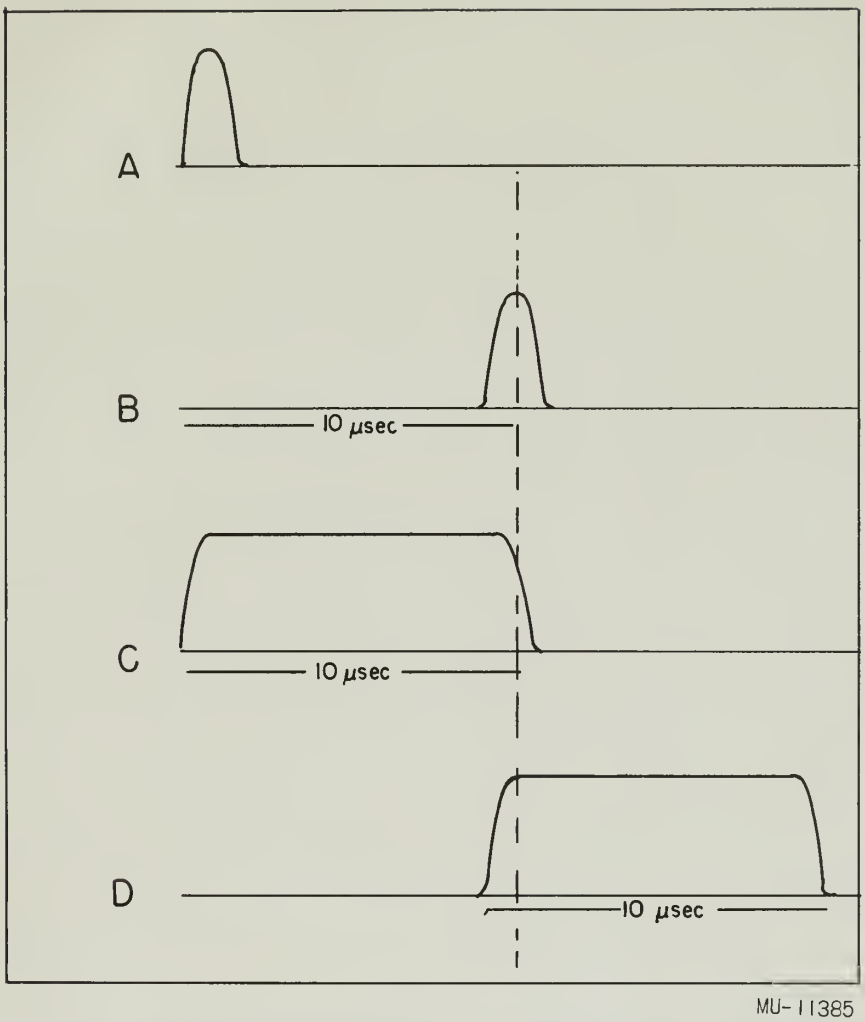

Fig. 7. Relative positions of the two extreme cases suitable for occurrence of a coincidence.

Curve A, photomultiplier pulse;
Curve B, photomultiplier pulse delayed 10 μsec;
Curve C, stretched pulse formed by BF_3 pulse occurring immediately following the PM pulse (A);
Curve D, stretched pulse formed by BF_3 pulse occurring 10 μsec after the PM pulse (A).

initially tested with a moderately strong Po-Be neutron source (5×10^6 n/sec), in order to check general counting characteristics, but in particular to learn the accidental-counting rate under different conditions. The accidental-counting rate can be calculated from the equation

$$ACR = N_1 N_2 \tau_1 \tau_2 \left(\frac{1}{\tau_1} + \frac{1}{\tau_2} \right) ,$$

where $N_1 + N_2$ are the singles-counting rates of the individual counters and τ_1 and τ_2 their respective pulse widths. With the counter unshielded and with the source placed so as to give a moderate BF_3 counting rate, the ratio of real counts to calculated accidental counts was at a prohibitive low of ~1.2. A shield of 2 inches of lead had a small beneficial effect, decreasing the gamma flux and thereby the total number of photomultiplier counts. A double shield of cadmium, including a tight-fitting jacket around the counter, and an outer closed box of 30-mil total thickness, increased the ratio of real counts to accidental counts to ~1.45, which was still an unacceptable ratio. Calculations using scattering and capture cross sections indicated that the probability that accidental thermal neutrons would enter the counter was extremely small, and that epithermal neutrons must be passing through the cadmium, and being captured. A 2-inch layer of B_4C powder (Norbide No. 20 grit) was placed around the counter in a cylindrical double-walled canister of 10-mil galvanized iron, and proved to be a successful shield, eliminating background to such an extent that the ratio of reals to accidentals was as high as 9.75.

CHAPTER III

RESULTS OF THE SPECTRUM ANALYSIS OF SEVERAL NEUTRON SOURCES

Figures 8 and 9 show the results of analysis of a Mock Fission source and of a Po-Be source with this spectrometer. The original intention of analyzing the Po-Be source was to obtain one or two calibration points for the counter by observing the positions of the known peaks at 2.8 and 7.3 Mev as measured by Richards.[13] In Fig. 9 it can be seen that the shape of the Po-Be curve is not inconsistent with that obtained by Richards. For this comparison, we used the flex point in the curve at ~55 volts and from this sketched in a second peak, which may be submerged in the low pulse background. In addition to this indication of a peak, there is a clearly resolved peak at ~91 volts. The ratio of peak heights in the known Po-Be spectrum is ~2/1 and the measured ratio is ~3/1. The ratio of peak energies is 2/1 in both the Po-Be spectrum and the measured spectrum. This is a very generous interpretation of the data, as will be seen by the response of the spectrumeter to a monoenergetic flux of neutrons of 14 Mev. The Mock Fission source, whose spectrum is known to have a peak at 1 Mev, was used in an attempt to discover if the counter would be useful in this range. The result, as seen in Fig. 8, was again not inconsistent with the previously measured spectrum.

In order to explain the observed spectra one can assume that the incident-neutron spectra have been shifted into the observed spectra seen in the figures, or that the spectrometer response is sufficiently nonlinear to account for the observed curves, or that there is a combination of these two effects. First, if sufficient scattering material were present between the spectrometer and the source of the neutrons, or around the spectrometer, the incident-neutron energy spectrum could be shifted toward lower energies. This scattering alone could result in the apparently observed build-up of the number of counts in the lower-energy regions, and simultaneously cause a partial or complete

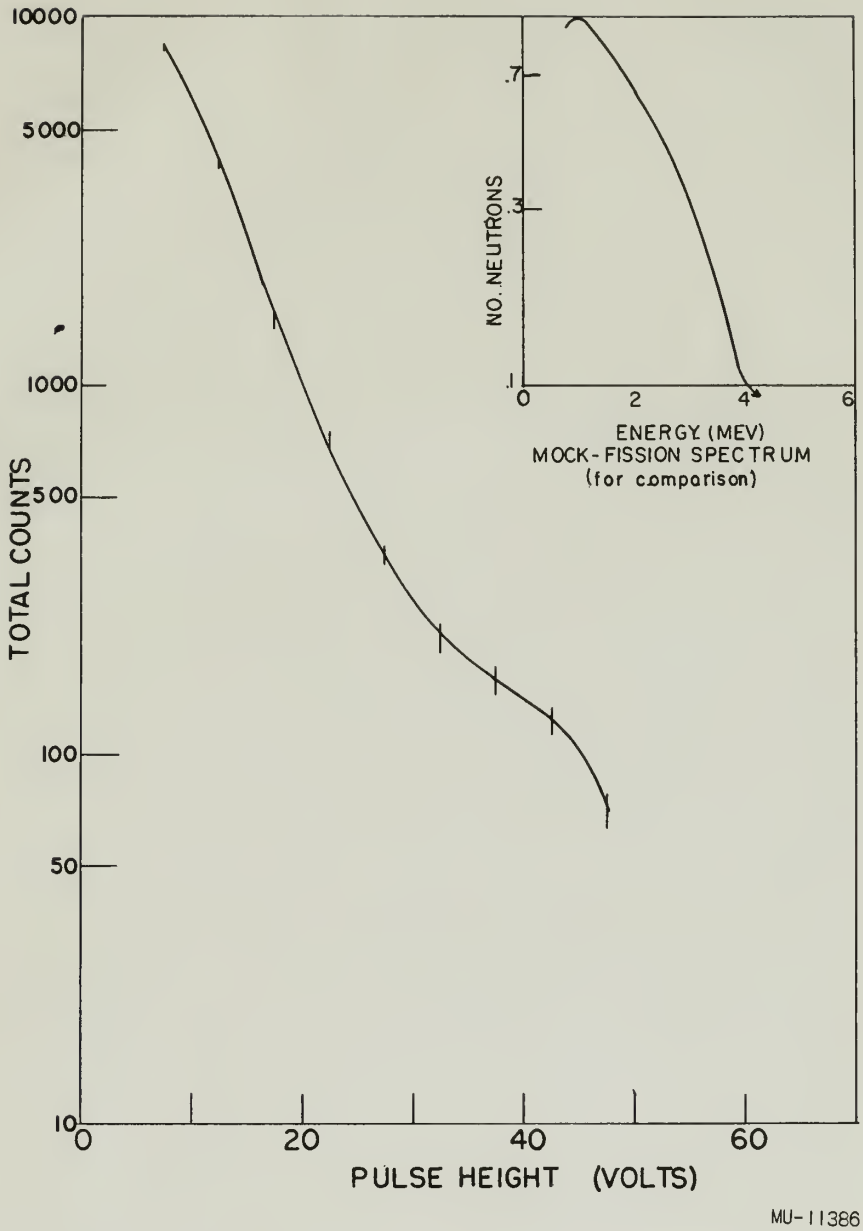

Fig. 8. Mock-fission neutron-source spectrum analysis.

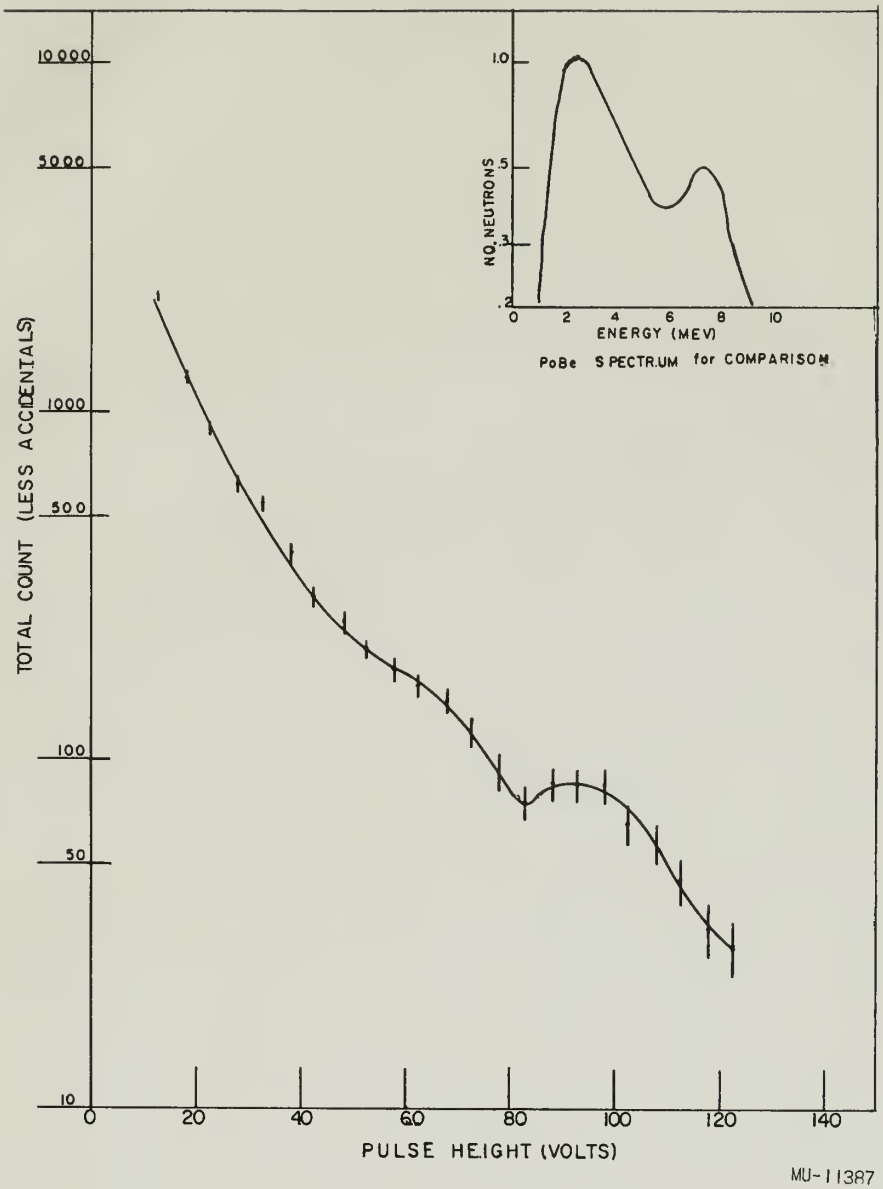

Fig. 9. Polonium-beryllium neutron-source spectrum analysis.

disappearance of any maxima. In the second place, the same result could be obtained if no scattering material were present, but the spectrometer were not able to resolve energies in the given range or had a serious nonlinearity. In addition, multiple scattering--resulting from more than one recoil from each neutron--combined with the known nonlinear response of the scintillation crystal itself, and with small instabilities in the pulse-height analyzer (particularly true at low counting rates which extend over several days), could serve to shift the observed pulse-height spectrum. The true behavior of the spectrometer probably lies between these alternatives, with increasingly poor resolution and loss of linearity for neutron energies below 3 Mev.

The spectrometer, as it was used in these analyses, utilized 2 inches of B_4C surrounding the BF_3 counter to stop thermal and epithermal neutrons of external origin that could be immediately captured by the BF_3 counter and thus increase the accidental counting rate. There was a 42% probability of a collision between the incoming neutrons (~3 to 10 Mev) and a boron atom, with an average energy loss of 15%. Future modifications should include a B_4C sheath only 0.5 inch thick, which should be adequate to stop the incident slow neutrons without appreciable moderation of the fast neutrons.

Figure 10 shows the results obtained with the spectrometer when an attempt was made to calibrate it with 14-Mev monoenergetic neutrons obtained from a $t(d,n)\,He^4$ reaction at the UCRL Livermore Cockcroft-Walton accelerator. The high flux of neutrons necessitated the placing of the spectrometer as far as possible from the target, which resulted in the undesirable geometry of having the counter on a concrete floor rather than suspended as far from reflecting surfaces as possible. Figure 11 shows the experimental setup and indicates the means of collimation used. A curve resulting from an ungated exposure of the counter to a 1-millicurie Co^{60} gamma source is also included in Fig. 10. This information is furnished for a comparison of the response of the scintillator to gamma radiation and recoil protons. In addition, it gives a calibration point for the pulse-height analyzer. As can be seen, if the

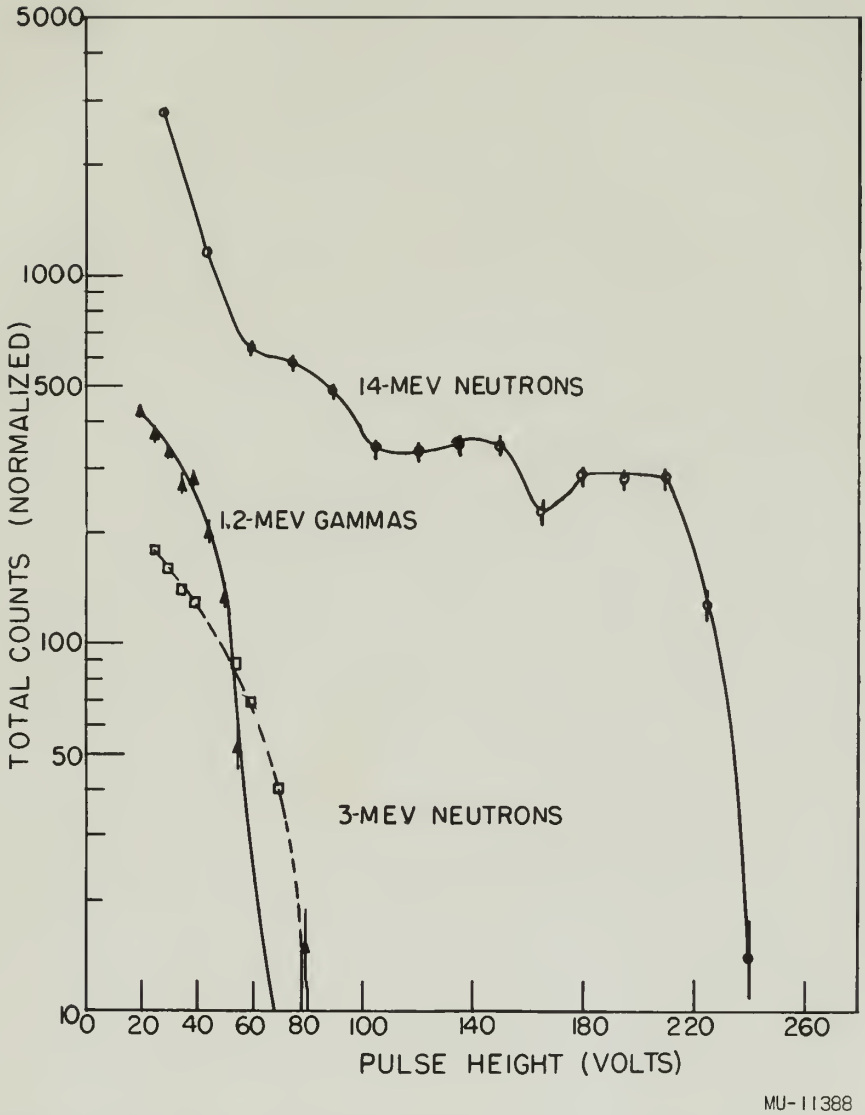

Fig. 10. Observed pulse-height spectrum from 14-Mev monoenergetic neutron source. [t(d, n)He^4] The 1.2-Mev gamma (Co^{60}) curve and the 3-Mev neutron curve (d, d) are included for comparison purposes.

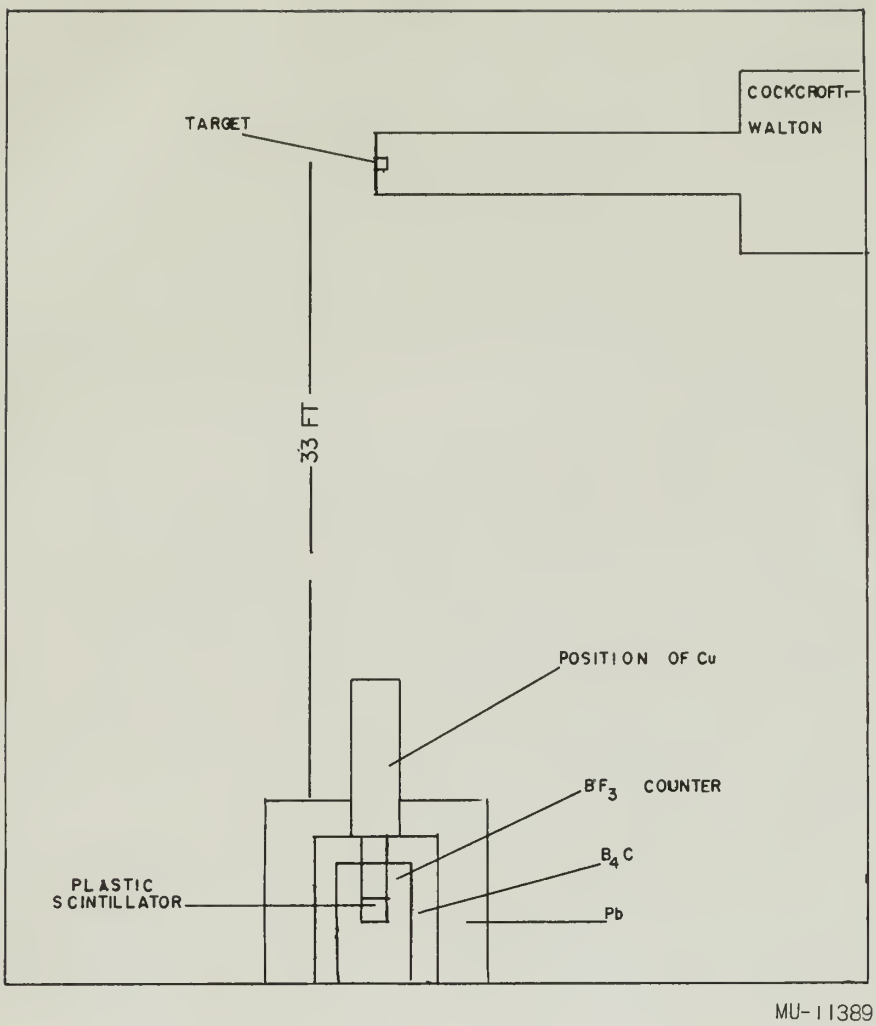

Fig. 11. Experimental setup during spectrometer exposure at the Cockcroft-Walton Accelerator, UCRL, Livermore.

maximum Co^{60} 1.2-Mev gamma-ray response is multiplied by 2, the energy scale will be approximately linear from the 2.4-Mev to the 14-Mev position, which agrees well with the relative pulse heights expected for gammas and protons of equal initial energy discussed in Chapter I.

In addition to the test for which the results are recorded in Fig. 10, a short run was made with a deuterium target substituted for the tritium target. This was in order to investigate the possibility that the effect experienced may have been due to the 3-Mev neutrons from a $d(d,n)He^3$ reaction occurring along the walls of the vacuum chamber and beam tube through which the accelerated deuterons travel prior to bombarding the target. It is known that as a bombardment proceeds, deuterons collect on these walls and act as secondary targets, and that relatively large numbers of 3-Mev neutrons are thus produced along with the 14-Mev neutrons. Using the deuterium target would eliminate all the 14-Mev neutrons. Owing to a shortage of available time, and low counting rate, the statistics are extremely poor. The pulses from the 3-Mev neutrons all lay below the 3-Mev point that was based on the location of the 14-Mev point in the pulse-height analyzer. The data indicate that the 3-Mev neutrons gave pulse heights of approximately the same size as the 1.2-Mev gammas.

At this time we can only surmise that the counts occurring between the noise peak at the left and the maximum height of the 14-Mev position are moderated neutrons not arriving directly from the target. To investigate this point a solid cylinder of copper, 30 inches long and 4 inches in diameter, was placed on a line between the target and the collimator for the spectrometer. The number of counts in the vicinity of what is apparently the 14-Mev neutron channel on the pulse-height analyzer dropped almost to zero, while the low-pulse-height channels continued receiving counts at approximately the same rate. The neutron spectrum arising from the $t(d,n)He^4$ reaction at the UCRL Livermore Cockcroft-Walton accelerator has been measured with a time-of-flight spectrometer placed close to the target. Owing to the technique of this measurement the spectrometer did not observe lower-energy neutrons

scattered from the walls of the concrete vault in which the target is located. The spectrum secured for this work shows a sharp peak at 14 Mev and very little between this and the 3-Mev $d(d,n)He^3$ peak. Our spectrometer, although it observed the same target and was somewhat shielded from the general background of neutrons in the vault, is inherently sensitive to neutrons arriving from a 4π solid angle, so that although the 30 inches of copper is able to reduce the direct 14-Mev neutrons by a factor of 42, it does not have much effect on the neutron atmosphere leaking into the spectrometer from the sides and top.

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