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Principles of Radioisotope Methodology

Gary Rothwell
Ouachita Baptist University

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Mr. Glen Good
By: Gary Rothwell
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Honors Project
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Introduction

This honors project was designed for familiarization with modern instrumentation and experimentation methods for radioisotope detection and energy calculations. The two main instruments I studied were the Geiger-Mueller Counter and a single channel pulse height analyzer. This report will include a brief history of radiation and a discription of the more important types. Next is a description of the two instruments mentioned and some simple experiments I conducted using them.

Historical Survey

Radioactivity was observed as early as 1867 when Niepce de Saint-Victor noticed a fogging of silver cloride emulsions by uranium salts. He reported that blackening of the emulsions occured even when they were separated from the uranium salts by thin sheets of paper. He did not recognize the true cause of the blackening (radioactivity) but attributed the effect to luminescence phenomena.

When Henri Becquerel repeated some of these experiments in 1896, he too believed that flurescence was associated with the fogging of his photographic plates. Becquerel did not realize either that he had observed the result of a process of spontaneous transmutation, akin to one of the long-sought

goals of the alchemist, and was surprised and puzzled to learn that photographic plates were still darkened by uranium salts even when the salts had received no previous "activation" with light. It was not until 1898, after Marie Curie had studied the radiation emitted by uranium, that a truly new phenomenon was recognized. The fogging of the photographic plates was now known to be caused by some sort of penetrating radiation similar to the x-rays of Roentgen. Produced not only by uranium and thorium salts, radiation was also observed from two new elements, polonium and radium, which had just been discovered by Marie Curie.

By 1899 Rutherford had demonstrated that the radiation emanating from uranium salts and from a variety of other salts and minerals was of two types, which he called alpha and beta. In 1900 a third type of radiation was observer be P. Curie and Villard; they called it gamma. It was only natural that much curiosity should be aroused concerning the origin--one must realize that it was not until 1908 that Rutherford and Geiger determined the charge of the alpha particle and not until 1909 that Rutherfordand Royds identified the alpha particle as a positively charged helium nucleus. It should be further realized that the nuclear theory, as taught even at the high school level today, was unknown until 1911 when Rutherford demonstrated by alpha-particle-scattering experiments that the positive charge of an atom must be concentrated at one point (the nucleus) rather than uniformly distributed throughout the volume of the atom as had been suggested be J. J. Thomson.

Rutherford's nuclear theory was the break-through which made possible a rapid series of developments in both atomic and

nuclear theory. For example, in 1912, J. J. Thomson, using positive-ray analysis (a forerunner of mass spectrometry) to measure particle mass, showed that two types of neon existed, with masses of 20 and 22. The following year these two forms of neon, differing only in mass, were separated by F. W. Aston by fractional diffusion through clay pipe stems, thus confirming the work of Thomson. F. Soddy proposed the name <u>isotope</u> (Gr; equal place) to denote a different mass of the same element.

By 1913 Niels Bohr had proposed his theory of atomic structure which applied the principles of Planck's quantum theory to Ruterford's planetary model of the atom. Proof of the Bohr theory was provided by spectral data, the utility of which is based on the principle that the energy associated with a particular line in the spectrum of an element is equal to the energy lost by an electron in passing from one specific orbit (energy level) to another in the Bohr model of the atom. Yet, the spectra which provided evidence for validity of the Bohr atom ultimately pointed to its shortcomings. Certain hyper-fine structures of the spectra cannot be explained by the Bohr theory.

Bohr had dealt with the atom from a particulate point of view, in which he assumed that particles called electrons revolve about a particle called the nucleus. In 1923, when Count Louis de Broglie suggested that the dualism of the wave and particle functions of radiation (waves versus photons) might apply to material particles as well, Schrodinger quickly applied the de Broglie theory to the atom. According to de Broglie, every particle in motion has associated with it a wavelength.

Based upon the wave function, the wave mechanical theory proposed by Schrodinger in 1926 differs considerably from the rather rigid mechanical model of Bohr but presents the most acceptable atomic model known today.

As our knowledge of the extranuclear structure of the atom was extended by persistent investigation, information about the nucleus, too, was gradually forthcoming. Thus Rutherford's investigations led to the very important observation of artificial transmutation first performed in 1919. In the years that followed many notable experiments were performed, many discoveries made, and many concepts proposed. These researches culminated in the discovery of fission, which led to the first atomic pile, constructed by E. Fermi in 1942 in Chicago. This development not only made possible production of the nuclear bomb, but also made practical largescale use of radioisotopes in research, medicine, industry, and in numerous other fields. Over 1300 species of atoms (nuclides) are presently known, and these may have an infinite variety of applications. It can be safely said that almost everything made today has at some point in its development or production been benefited by the use of isotopes.

Types of Radiation

The emanation of rays by radioactive substances is caused by the random disintegration of their inherently unstable nuclei into more stable nuclei. The rays given off

by a radioisotope may be of the following three types (or mixture of the three):

Alpha rays (α) -- These are really particles. They are positively charged particles which are identical to helium ions but differ in that they originate from the nucleus of an isotope. They have great ionizing power and travel about 0.1 the speed of light.

Beta rays (β) -- These are negatively charged particles which are identical to electrons except that they originate in the nucleus. They have much less ionizing power than α particles and travel up to 0.9 the speed of light. Their range is much greater than α particles.

Gamma rays (γ) -- these are rays similar to X-rays but of shorter wave length. They have very low ionizing power, but very great range. They travel at the speed of light.

Other types of nuclear changes may occur with other types of radiation given off. Less common types of radiations emitted are neutrons and positrons. The neutron needs no explanation, and the positron is identical to the beta particle (negatron) except for the positive charge of the positron.

Of all the types of radiation enumerated above, only the neutron can cause a substance to become radioactive. For this reason neutron-emitters are regulated stringently by the authorities. Exposure to or the handling of alpha, beta, or gamma-emitting radioisotopes does not cause any radioactivity.

Geiger-Muller Counters

A Geiger-Muller counter is a device used for the detection and measurement of radiation. Basically, it consists of a pair of electrodes surrounded by a gas especially selected for the ease with which it can be ionized. When radiation ionizes the gas, the ions so produced travel to the electrodes between which is maintained a high electrical potential. The motion of the ions to the electrodes constitutes an electric current which is detected and recorded by a scaler. Thus, each particle or ray of radiation passing through the Geiger-Muller tube causes a short pulse of current to flow, the number of such pulses being a measure of the intensity of the radiation.

Geiger-Muller counters are supplied in a variety of forms. A typical "end-window" type of tube is depicted in figure 1. This tube is so named because it has a thin window at one end through which the radiation passes. It consists of a metal or glass cylindrical envelope the inside of which has been coated with a conducting material. The wall of the tube constitutes the negative electrode known as the cathode, In the center, concentrically aligned, is a fine wire which serves as the anode. It is charged positively, often to approximately 1200 volts, with respect to the cathode.

The space between the electrodes is filled with a gas, helium or argon usually being used. The window prevents the escape of the gas to the atmosphere, yet is sufficiently thin so that it does not prevent the passage of radiation into the tube to any appreciable extent, especially the passage of beta

particles for which this type of tube is most useful.

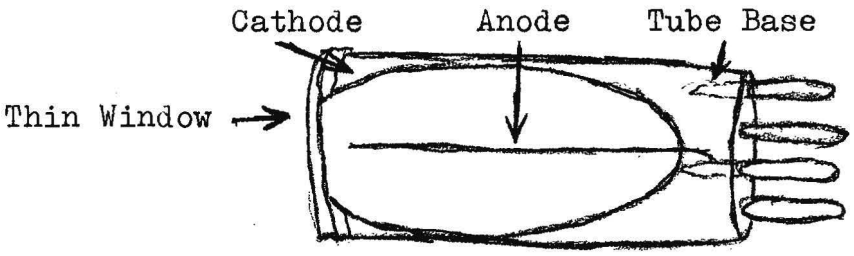


Figure 1

A beta particle entering the counter produces a number of ion pairs consisting of electrons and positively charged ions of the gas filling the tube. Under the influence of the electrical field between the electrodes, the electrons travel to the center wire or anode. In the process, however, they themselves acquire enough energy that their collisions with gas molecules result in the formation of still other ion pairs. This process continues with the resulting formation of an avalanche of electrons and positive ions. The positive ions travel toward the outer envelope, the cathode, also causing additional ion pairs to be produced. Thus, once ionization is initiated, the tube would continue to discharge continuously unless turned off or quenched by some other process.

One way to quench a tube in order to restore it to its original quiescent state is to remove the high voltage momentarily. This can be done electronically and is called external quenching. Another method more commonly used today is to employ internal quenching by mixing a small quantity of a polyatomic gas with the counter gas to absorb some of the energy of the electrons and positive ions after an ionizing

event. In the process the polyatomic molecule is decomposed. If a substance like alcohol or butane is used as a quenching agent, the tube is said to be "organic-quinched". Such a tube has a useful life of about 10^8 counts because the molecules are decomposed irreversibly. "Halogen-quenched" tubes utilize chlorine, bromine and their compounds, as quenching agents. These tubes have a much longer life because the atoms normally recombine.

The net behavior of a Geiger-Muller tube during ionizing is the result of two opposing groups of factors, those tending to perpetuate discharge and those tending to limit discharge in the preceding paragraphs.

Figure 2 illustrates a typical characteristic curve for a Geiger-Muller tube.

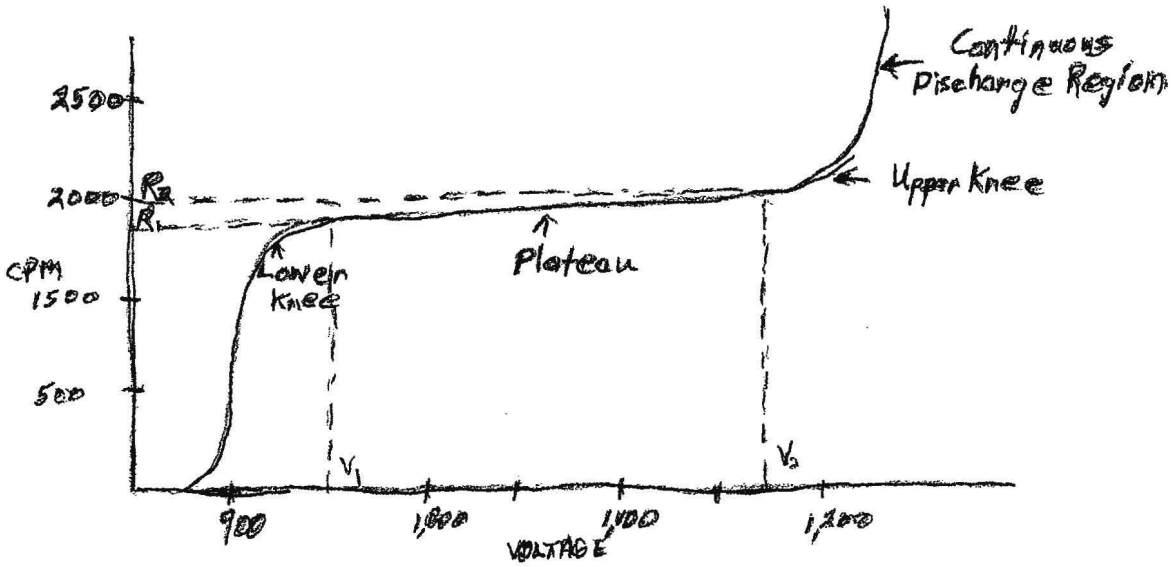


Figure 2

Characteristic Curve for Geiger-Muller Tube

If a sample is placed beneath a tube and the voltage which is impressed on the tube slowly increased, a voltage will be reached at which the G-M tube just begins to perceive a few counts as indicated by the scaler. This is the starting potential. Now as the voltage is increased very slightly a very rapid increase in the counting rate is observed. This voltage is known as the threshold. Beyond the threshold, further increases in the voltage over quite a range will produce little effect on the counting rate. This region, known as the plateau, should have a slope of less than 3% for good tubes. Within the plateau region the proper operating voltage is selected. The operating voltage should be selected relatively close to the threshold voltage (within the lower 25% of the plateau) to help preserve the life of the tube. Also the operating voltage should be selected at a point where the plateau shows minimum slope. If the voltage is indiscriminately increased beyond the plateau region, the region of continuous discharge is reached and the tube may be seriously damaged.

The plateau slope is of value since it serves as a figure of merit for a counter tube. The slope $\Delta R/\Delta V$ defined using the notation of figure 2 as simply $(R_2 - R_1)/(V_2 - V_1)$ is meaningless because the merit of the tube could be "improved" through the simple expedient of reducing the sample activity. The normalized plateau slope is used as the figure of merit. It is calculated as the percentage change in counting rate R divided by the percentage change in applied voltage V using the threshold values as the base.

$$\frac{100\,(R_2-R_1)/R_1}{100\,(V_2-V_1)/V_1} = \frac{(R_2-R_1)\,V_1}{(V_2-V_1)\,R_1} = m$$

Very often the slope is expressed as the per cent increase in counting rate per volt (or per 100 volts) increase in the applied voltage. Thus the relative plateau slope is expressed as

$$m = \frac{100\,(R_2 - R_1)/R_1}{V_2 - V_1} \qquad = \text{per cent per volt}$$

$$m = \frac{100\,(R_2 - R_1)/R_1}{V_2 - V_1} \times 100 \qquad = \text{per cent per 100 volts}$$

A good tube should have a slope of less than 10% per 100 volts. often the slope is as little as 3% per 100 volts.

After a Geiger-Muller tube has been exposed to a high intensity gamma source the background of the tube may be abnormally high for some time after the source has been removed. Once ionization of a Geiger tube has been initiated, the tube becomes insensitive for a short interval of time. This interval is called the resolving time. It represents the time during which two or more ionizing particles striking the sensitive portion of the tube will be counted as a single particle. As a result of this phenomenon, the number of counts recorded will be less than the actual number of particles passing into the tube. The difference between the true and observed count is known as the coincidence loss.

Geiger tubes are not equally sensitive to alpha, beta and gamma radiations. This is explained by considering both the properties of the radiation and the properties of the Geiger tube. To initiate discharge of the tube, the radiation must first reach the sensitive volume. Alpha particles, being the least penetrating, may be absorbed by the window unless it is very thin. Beta particles are more penetrating and gamma rays are very penetrating. Thus thicker windows can be tolerated

with the latter two types of radiation. It is found that the efficiency of the Geiger tube is essentially 100% for alpha particles, nearly 100% for beta particles but only 1 or 2% for the gamma radiation. They are so penetrating they pass on through without ionizing the gas.

Below is the data for the graph on the next page.

Source Cs 137
Background (V=1000)
533/10 min
476/10 min

Voltage	Counts/Min
600	0
650	12
700	968
750	1737
800	3080
850	3429
900	3615
950	3803
1000	3900
1050	4098
1100	4263
1150	4455
1200	4452
1250	4938
1300	8690
1350	11645

M Sound Grown

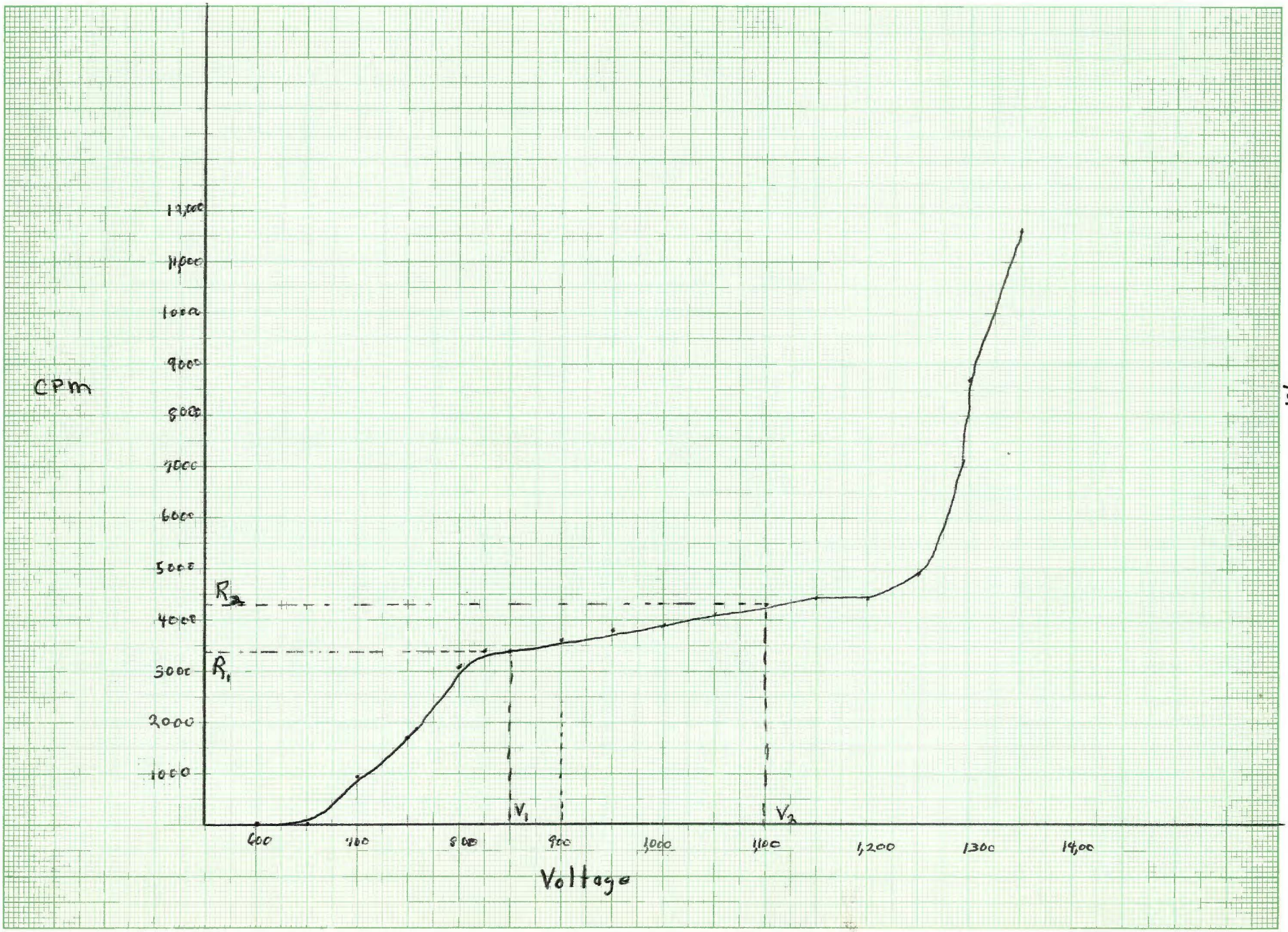
$V_1 = 900 \qquad R_1 = 3615$

$V_2 = 1100 \qquad R_2 = 4263$

$$m = \frac{(4263-3615)\,900}{(1100-900)\,3615} = .758$$

$$m = \frac{100(4263-3615)/3615}{1100-900} \times 100 =$$

$= 8.962$ per 100 volts



Characteristic Curve for Geigen - Mullen Tube

Another experiment I did to gain use of the Geiger-Muller tube is to determine the effect of distance upon radiation. Using the same source and background, I obtained the following data. (I make use of the shelf number instead of the actual distance because the shelves are evenly spaced from one another.

Relative Distance (Shelf number)	Activity (c/m)
1	42,691
2	23,063
3	11,249
4	6,092
5	3,800

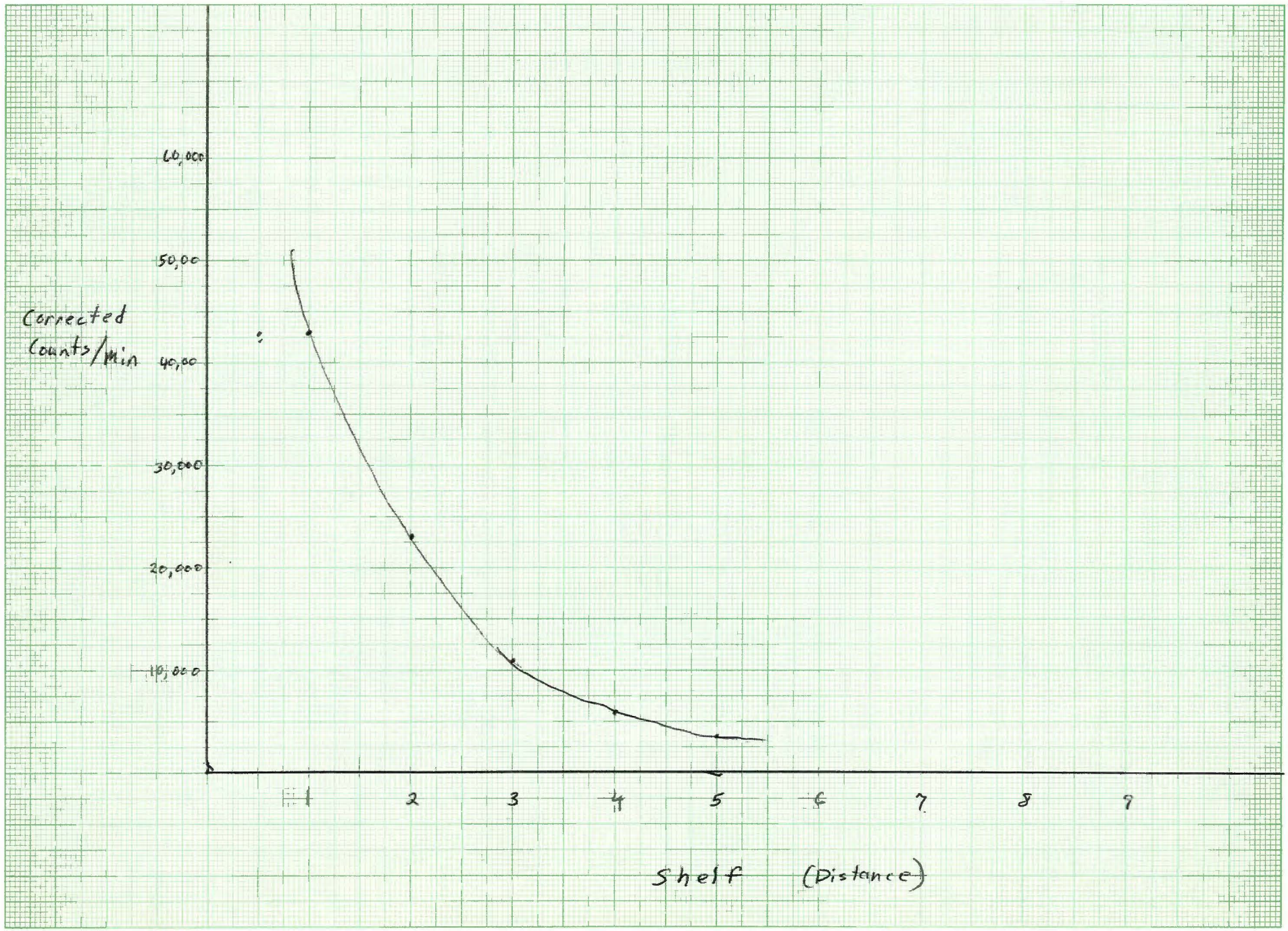
(Subtracted 50 c/m from each reading for background radiation)

From this graph we can see that the amount of radiation decreases with the increase in distance. This looks to be exponentially.

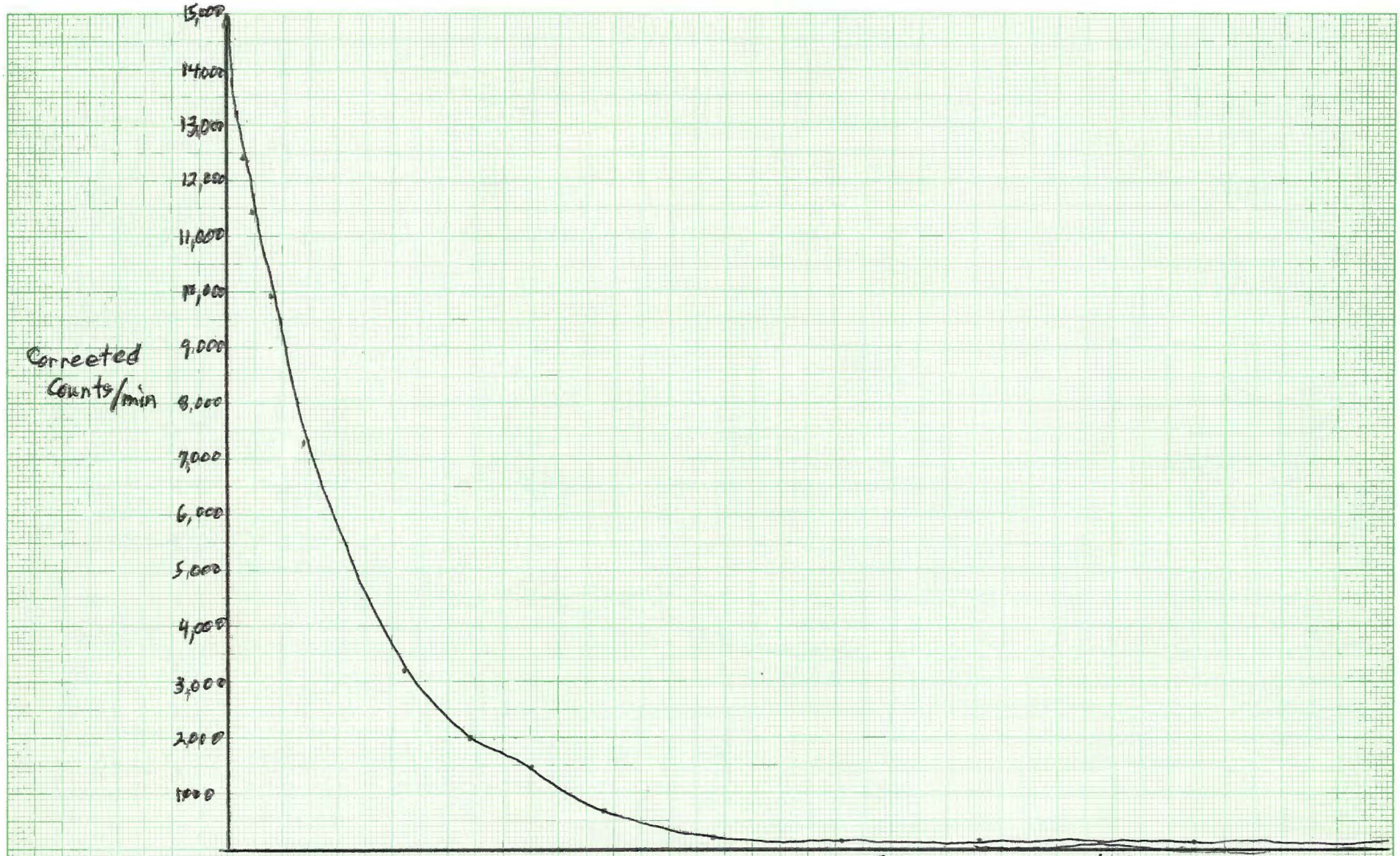
One final experiment using the Geiger-Muller tube counting apparatus was to determine the absorption of beta and gamma radiation. In order to do this I used different thicknesses of aluminum and lead. The data follows with the graph on page 15.

Background - 46c/m Source - Cs 137

Absorber thickness (mg/cm^2)		Corected counts (c/m)
0		14,865
3.0		13,217
6.7		12,362
13.6		11,466
20.3		9,863
34.5		7,269
82.2		3,237
111.4		2,048
135.5		1,505
171.7		739
222.3	Aluminum	242
277.3		165
342.6		155
436.5		147



Effect of Distance Upon Radiation



Asorption of Beta + Gamma Radiation

Source - Cs 137
Background - 45
Absorber - Al

The graph on page 15 shows a quick drop then a leveling off of the count around 150 c/m. This is due to the asorption properties of beta and gamma radiation. The beta rays account for the fast drop and due to the penetrating ability of gamma radiation it levels off around the thickness 250 mg/cm^2. I also took the same type data for lead asorption but will not include that redundant graph due to lack of data of different widths.

Background - 49c/m Source Cs 137

Absorber thickness (mg/cm^2)		Corrected Count (c/m)
925.3		168
1821.3		158
2650.7	Lead	139
4493.6		129
7140.8		110

Single Channel Pulse Height Analyzer

The scintillation counter operates on the principle that certain materials, called phosphors, on exposure to radiation, convert the kinetic energy of the particle or photon of radiation to flashes of light (scintillations).

Scintillation counting was one of the first known methods for detecting nuclear radiation and determining the activity of a radioactive sample. However, it has not been widely used until recently along with improved detectors and electronic counting techniques. At the time of Rutherford's scattering experiments in 1911 scintillation counters were used but the flashes of light had to be detected by the human eye and counted manually.

The instrument used in my experiment consisted of a detector, an analyzer and a scaler or rate meter. The detector consists of a scintillation detector (crystal of NaI activated with Tl), through the crystal (Shielded from light on the outside and in optical contact with the PM) a small flash of light is emitted, the intensity of the light being proportional to the energy of the gamma ray. The photomultiplier "sees" the flash of light, converts it into a pulse of electricity and multiplies the pulse to a measurable quantity. This pulse is then amplified further by the preamplifer. The voltage of the pulse that passes to the analyzer is proportional to the energy of the gamma ray, or radiation.

The analyzer consists of a linear amplifier, two discriminator circuits and a voltage regulator. The gain of the amplifier can be set by the gain control. The data obtained on one gain setting, with interpretation, may be used along with data obtained on another gain setting but this is not desirable. The gain setting determines the multiplication of the voltage of the pulse height coming to the analyzer. This amplified pulse goes to the discriminator circuits.

All scalers and count rate meters use an electronic circuit called a discriminator. As indicated by its name, the discriminator circuit responds only to voltage pulses above a certain minimum value. In a G-M counter scaler, the discriminator's sensitivity can be adjusted from one-fourth to ten volts, and is usually set at the lower value. By means of this circuit, electrical noise pulses can be rejected and only the larger G-M pulses counted.

In the spectrometer which I used in this experiment two discriminators were used. One discriminator is set at a fixed voltage above the other. A common control is used to adjust the sensitivity of both discriminators together (this control being marked Base on the analyzer). By means of the Base Control, the sensitivity of the lower discriminator is variable from zero volts (approximately 0.1 V) to 100 volts. The sensitivity of the upper discriminator is set at 0.1 volt to 10 volts higher (as determined by the Window Control). Let us assume that it is set two volts higher. Then the Base Control would vary the sensitivity of the upper discriminator from 2 to 102 volts, corresponding to the 0 to 100 volts for the lower discriminator.

If the Base control is set at 20 volts and the window is set at two volts, a 21-volt pulse would trigger the lower discriminator, but not the upper. In like manner a 22.1 volt pulse would trigger both discriminators and a 19.9 volt pulse would not trigger either one. The upper discriminator can be used to close an electronic gate and the pulse from the lower discriminator can be delayed (approximately 1 microsecond) until the gate is closed if the upper discriminator is triggered. With such an arrangement, the 21-volt pulse would trigger the lower discriminator and the output pulse would pass through the gate to be counted since the upper discriminator did not close the gate. However, the 22.1-volt pulse would cause the upper discriminator to close the gate so that the pulse from the lower discriminator could not pass through and therefore would not be counted.

By such a system, we could set the Base control at 10 volts and count those pulses that were between 10.0 and 12.0 volts. Then, readjusting of the Base control would let us count those pulses between 12.0 and 14.0 volts. We can obtain a distribution of relative numbers of counts per two-volt pulse height increment versus pulse height in volts by moving the Base Control from 0 to 100.

Since the scintillation counter exhibits a great dependence of the counting rate on the applied voltage an additional voltage regulator that can be set to the desired voltage is included in the analyzer.

In use when the base voltage is varied and the activity of a particular sample is measured the activity will remain

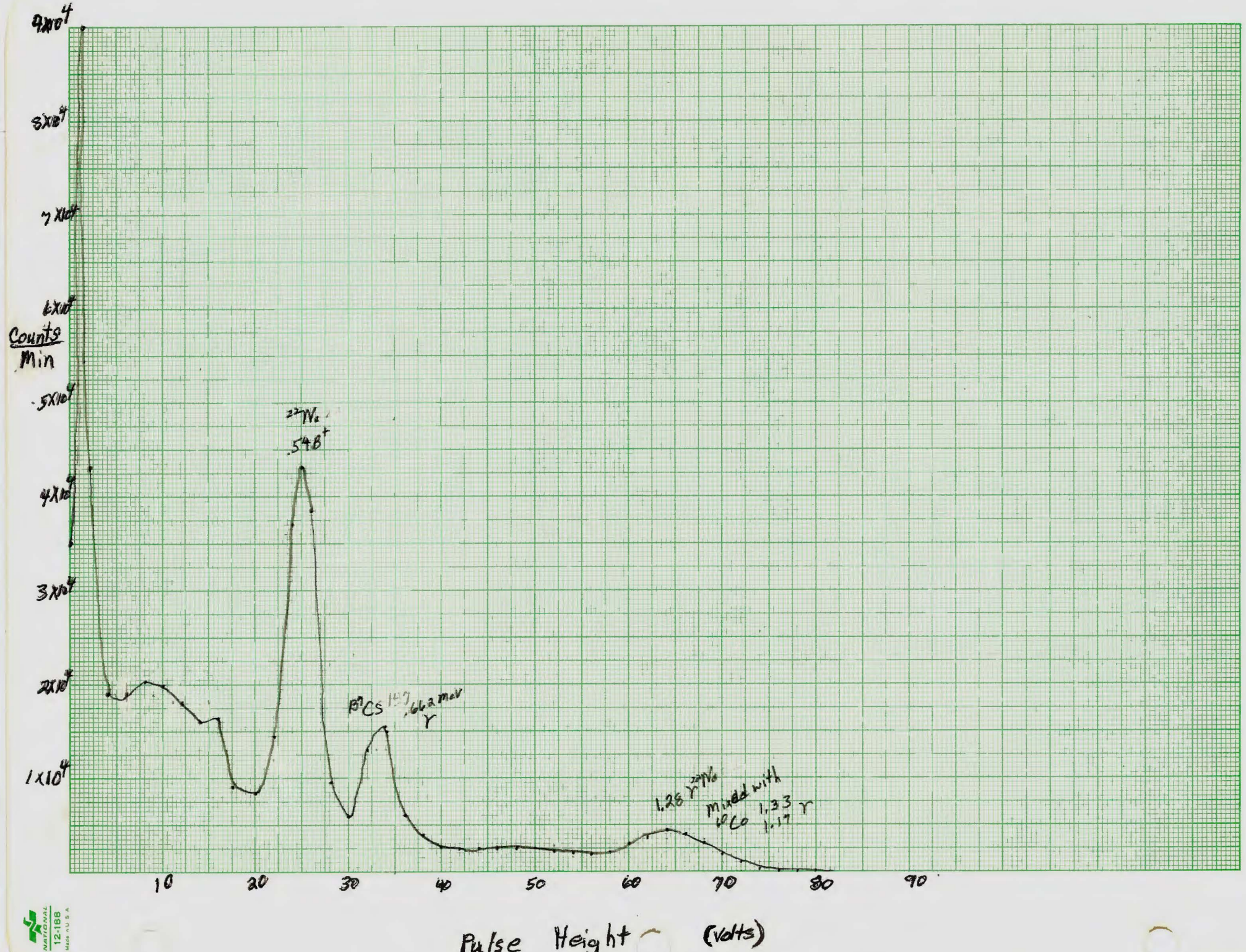
relatively low until the voltage is reached which allows the pulse from the gamma rays being counted to register on the scaler or rate meter. Thus in a plot of cpm vs base voltage there should be a peak for each gamma ray emitted. This type of measurement can be made by using isotopes which have known gamma ray energies as a calibration. In such a calibration, a plot of the voltage, at which the peak has its maximum vs the gamma ray energy should give a straight line. Thus if the position of the peak of an unknown gamma energy is known its energy can be read from the calibration graph.

For the last part of my project I used this pulse height analyzer to try and graph the energies for three different samples combined (^{60}Co, ^{22}Na, and ^{137}Cs). The data and graph are on the two preceeding pages. With the graphs of the individual elements I determined what I thought the labled radiation plots are. I believe that the ^{60}Co source was too weak to show up unless it just helped to spread the 1.28 γ ^{22}Na out. This could be checked by running a plot of each of these elements using the same settings on the analyzer. This was not done due to lack of time.

Ratemeter high voltage = 1,500 volts
Analyzer high voltage = 820 volts
Sources ^{60}Co, ^{22}Na, ^{137}Cs

Gain 4 (Double pulse height of the peak to find rough estimate of energy in 10^{-2}mev's)

Pulse Height (Volts)		Counts/Minute (10^2)
0		350
1.3		900
2		428
4		190
6		190
8		202
10	(odd values at peaks)	198
12		182
14		160
16		162
18		90
20		85
22		145
24		370
25		430
26		360
28		95
30		58
32		130
33.7		155
34		150
36		60
38		30
40		25
42		25
44		25
46		25
48		25
50		25
52		23
54		21
56		18
58		21
60		30
62		35
64		45
66		40
68		30
70		18
72		10
74		2
76		1
78		0.5
80		0



Conclusion

This honors project has been worthwhile in two aspects. One I have accomplished my goal of learning how to use and getting some practical use of the instruments that I might come in contact with at graduate school in a nuclear laboratory. Second, as a result of using these instruments I learned more about radiation and applied what I already knew to some practical situations.

This was a really beneficial honors project and should be suggested to future physics majors or put into the physics requirements.