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Author
Anger, H.O.

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H. O. Anger

February 14, 1951

Berkeley, California

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Radiation Laboratory and Division of Medical Physics
University of California, Berkeley, California

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Introduction

During the few years since the papers of Kallman[1] and Deutsch[2] on scintillation counting appeared, many investigators have been working in the field. Phosphors for scintillation counting are now commercially available,[3] and a phototube especially suited to scintillation counting, the RCA 5819, is also available. In this laboratory we have attempted to apply scintillation counting to some of our problems in biological tracer research.

The principal advantage of scintillation counting from our point of view is that it makes possible the detection of gamma-radiation with much greater efficiency than is possible with G-M counters. This opened the possibility that we might be able to measure the activity of some of our biological samples by counting gamma-rays instead of beta-particles as we had been doing previously. Samples containing Fe^{59} or Co^{60} are examples. Both of these isotopes emit beta-particles of relatively low energy. They each emit two gamma-rays with energies of about 1.1 and 1.3 Mev. In the past we have counted the beta-particles from these samples with good efficiency but rather poor reproducibility because of the involved chemical procedure through which the biological tissue samples had to be put in order to prepare them for counting. The procedure consisted of chemically digesting the tissues to get them into solution and then electroplating the iron or cobalt content on metal discs in order to get samples that were thin enough so that the self absorption of the beta-particles was low. The use of scintillation counting makes it possible to eliminate the electroplating and other chemical preparation. The activity of the samples is measured simply by placing the tissues in glass vials without any other preparation and then placing the vials in the counter. The gamma-rays penetrate the tissue and the walls of the vial without appreciable loss and are counted when they are absorbed in the phosphor of the scintillation counter.

The counting efficiency of the scintillation method described here is about equal to the counting efficiency when beta-particles from electroplated samples are counted by means of a mica window G-M counter. The background count, however, is several times higher, making it impossible to count very weak samples. In spite of this, the scintillation method saves so much work in preparing the samples and gives such reproducible results compared to the beta-particle counting method that it is greatly preferred in this laboratory for tracer work with Co^{60} and Fe^{59}. It will no doubt prove equally useful for many other gamma emitting isotopes.

The Existence of a Counting Plateau

An important requirement of a counter for radioactive samples is that it must be adequately stable. At first it was thought that this might be a difficult requirement to meet in a scintillation counter since a stable counter would be very difficult to construct if a counting plateau similar to that of a G-M counter did not exist. We have found that a reasonably satisfactory plateau does exist when counting gamma-rays with energies of about 0.3 Mev and greater when a suitable phosphor and a selected 5819 phototube are used at room temperature. Anthracene, stilbene, and thallium activated sodium iodide are satisfactory phosphor materials. Calcium tungstate, cadmium tungstate, and terphenyl dissolved in m-xylene are materials from which we have not obtained good plateaus. However, Mayneord[4] has reported obtaining a plateau from calcium tungstate and an RCA 1P21 photomultiplier at liquid nitrogen temperature.

A typical plateau curve for a counter consisting of a large flat anthracene crystal and an RCA 5819 phototube is shown in Fig. 1. The crystal was quite large, about 1 3/4 x 1 3/4 x 7/8 inches, but contained many flaws. The flat side was cemented to the phototube face with transparent cement. The rest of the crystal was covered with aluminum foil to reflect light from the crystal into the phototube. The assembly was then covered with black photographic tape to exclude ambient light from the phototube.

Plateau curves are shown for gamma-rays from Co^{60}, Fe^{59}, and I^{131}. Unlike a G-M counter, the plateau slope is different for each isotope. If an operating voltage of 1850 volts is selected, the slope of the plateau is 4 percent per hundred volts for Co^{60}, 7 percent per hundred volts for Fe^{59}, and 10 percent per hundred volts for I^{131}. As will be explained later in this paper, the operating voltage given on these curves is not the actual voltage on the phototube. It is the voltage read on the scaler voltmeter, which is about 56 percent greater than the phototube voltage.

The I^{131} plateau is observed to have a steeper slope than the Fe^{59} plateau, and further more, it begins at a higher voltage. This is due to the fact that I^{131} emits gamma-rays of lower energy which produce weaker scintillations. If gamma-rays of still lower energy were to be counted, a shorter and steeper plateau would result and this method of counting would become less practical.

It may be wondered why the plateau slope for Co^{60} is less than the slope for Fe^{59} while the energy of the two gamma-rays emitted by each isotope is almost exactly the same. The reason is that Co^{60} emits its two gamma-rays in coincidence while Fe^{59} does not. Quite often, therefore, Co^{60} produces two scintillations in coincidence. This results in a bright scintillation that is recorded as a single count over a wider range of phototube voltage. The net result of these coincident scintillations is to give a longer plateau with slightly less slope.

The curve labeled background in Fig. 1 was taken with the samples removed, but with the crystal still in place, and therefore shows the background due to stray gamma- and cosmic-radiation and noise pulses from the phototube. The curve labeled phototube noise was taken with the crystal removed and therefore shows the contribution to the background of phototube noise alone. The background with the crystal in place and with 1 1/2 inches of lead shielding is 125 counts per minute at the operating voltage of 1850 volts. The contribution to this background from phototube noise alone is less than one count per minute at this voltage. Virtually the entire background is due to stray radiation entering the crystal.

The slope and length of the plateaus obtained depends not only on the type of phosphor and optical arrangement, but also on the sensitivity of the particular 5818 photoube used, a less sensitive tube giving a shorter plateau with greater slope. The tubes used to obtain the curves given in this paper were the best of a group of eight phototubes.

The resolving time of one of the above counters was measured and found to be about 6 microseconds. This corresponds to the resolving time of the scaler, as would be expected, since the decay time of anthracene and stilbene have been measured to be a fraction of a microsecond.[5] The decay time of thallium activated sodium iodide is approximately 0.25 microsecond.[6]

From the data presented, it seems that a very practical sample counter can be made which operates at room temperature. The existence of the counting plateau indicates that a counter with stable characteristics can be constructed. The fact that the count drops practically to zero when the crystal is removed indicates that phototube noise is not being appreciably counted. The background can be reduced further only by shielding the crystal and phototube or by some other method that will reduce the number of unwanted scintillations. Therefore, it is not necessary to refrigerate the phototube or to use two phototubes which count only coincidences in order to count fairly high energy gamma-rays with the three phosphor materials and the phototube mentioned above. However, such measures may still be necessary for counting very low energy gamma-rays. Other experimenters have reported on these methods.[2,7,8]

The Vial Sample Counter

As mentioned before, we have found it advantageous to count samples which are contained in glass vials. A drawing showing the general form of a counter of this type is shown in Fig. 2. The crystals are arranged around a well in which the sample vials are placed. Thus the counting geometry is high since the phosphor partially surrounds the sample. The aluminum housing serves as a light tight shield to prevent ambient light from entering the phototube. It also provides a reflecting surface for the scintillation light, and prevents stray beta-particles from entering the crystal. The brass absorber which fits between the vial and the aluminum housing provides part of the absorption path to prevent beta-particles from entering the phosphor from the sample, and also prevents the housing from becoming contaminated by activity that may be picked up from contact with the sample vials. If the absorber becomes contaminated from contact with the samples, it can easily be removed and cleaned.

We have constructed counters of this type using anthracene, stilbene, and thallium activated sodium iodide crystals. The most satisfactory counters were those which used thallium activated sodium iodide. They had the highest efficiency by almost a factor of three, and they also had the longest plateaus with the least slope. The anthracene and stilbene counters had plateaus which were satisfactory only for the higher energy gamma emitters.

The plateau curve for a typical vial sample counter with thallium activated sodium iodide as the phosphor is shown in Fig. 3. At an operating voltage of 1800 volts, the slope of the plateau is 2 percent per hundred volts for Co^{60}, 3 percent per hundred volts for Fe^{59}, and 6 percent per hundred volts for I^{131}.

The absolute counting efficiency when counting 2 milliliters of solution contained in a 1 dram glass vial is 29.8 percent for Co^{60} and 17.5 percent for Fe^{59} where the absolute counting efficiency is defined as the sample counting rate recorded by the counter divided by the disintegration rate of the sample. The background count when shielded by 1 1/2 inches of lead with an additional 1/4 inch of aluminum next to the counter is about 220 counts per minute. When shielded by about 4 inches of lead with 2 inches of steel next to the counter, the background is about 160 counts per minute. The background when the counter is unshielded is about 4000 counts per minute. The importance of at least a small amount of shielding for this type of counter is shown by the large reduction in background obtained with either of the lead shields.

Comparing this counter to a thin window G-M counter which counts beta-particles from electroplated samples, the absolute counting efficiency for Fe^{59} is 17.5 percent for the scintillation counter and about 16 percent for the G-M method. The backgrounds are about 160 and 25 counts per minute, respectively. Therefore, the scintillation counter is less sensitive by a factor of six for Fe^{59}. It is less sensitive by a factor of three for Co^{60}. However, larger samples can sometimes be used or longer counts taken to reduce the disadvantage for very weak samples.

The phosphor was made from two single crystals shaped and fitted together to form an annular ring with an outside diameter of 1 3/4 inches, an inside diameter of 3/4 inch, and a height of 7/8 inch. Since sodium iodide is hygroscopic, the crystals are protected from air by being immersed in <u>Nujol</u>, a clear mineral oil, after the method described by Hofstadter and McIntyre.[9] First, however, the yellow colored film which forms on sodium iodide crystals when they are exposed to air was removed. One way to remove the film is to immerse the entire crystal for about a minute in acetone, and then dry it and immerse it in Nujol. The acetone dissolves the yellow film and also some of the surface of the crystal leaving it very transparent. The crystals are then put into the aluminum holder and it is filled with Nujol. Then a lucite disc is fitted into the bottom of the aluminum holder to retain the oil. The side of the lucite disc nearest the phototube is provided with a curved surface which fits closely against the top of the phototube. A few drops of Canada Balsam between the lucite disc and the phototube provides a better optical path for the scintillation light. The two parts are then taped together with black photographic tape to complete the counter.

Fig. 4 shows a curve of the efficiency of the counter versus the volume of sample contained in the vial. This curve is of use when counting liquid or soft tissue samples of various volumes. The efficiency remains approximately constant until the volume reaches 1.5 milliliters, after which the efficiency begins to decrease. It is interesting to note that samples which occupy 1.5 milliliters or less can be counted at a known absolute efficiency without knowing the exact volume of the sample. Thus it is possible to measure the activity of small bits of biological tissue by simply packing them into the bottom of the vial and counting them. It may be necessary to homogenize the samples for the most accurate measurements if the activity is very unevenly distributed in the sample.

The plateau curve for a vial sample counter using anthracene crystals is shown in Fig. 5. At an operating voltage of 1800 volts, the slope of the plateau is 9 percent per hundred volts for Co^{60} and 11 percent per hundred volts for Fe^{59}. The slope of the I^{131} plateau is 34 percent per hundred volts when the counter is operated at 1900 volts. The increased slope of the plateau curves of this counter is partly due to the fact that it employs larger crystals than the one just described, and thus has lower optical efficiency. The crystals in this counter were shaped to form an annular ring with an outside diameter of 1 3/4 inches, an inside diameter of 3/4 inches and a height of 1 5/8 inches. The bottom of each crystal was cemented to the phototube window with Canada Balsam to obtain the best optical efficiency.

The absolute counting efficiency of this counter when counting 2 milliliters of solution contained in a 1 dram glass vial is 13.1 percent for Co^{60} and 7.0 percent for Fe^{59}. When counting 4 milliliters of solution in the same size vial, the efficiency is 11.0 percent for Co^{60} and 6.4 percent for Fe^{59}. The background count when shielded by 1 1/2 inches of lead with an additional 1.4 inch of aluminum next to the counter is about 120 counts per minute. When shielded by 4 inches of lead with 2 inches of steel next to the counter, the background is about 80 counts per minute. The background with the counter unshielded is about 600 counts per minute.

While the plateau slopes for Fe^{59} and Co^{60} are not quite as flat as those from a good G-M counter, this counter is still satisfactory for the higher energy gamma emitters since it is possible to control the high voltage from a good scaler to within 10 volts, and any error due to variation of the high voltage is therefor kept to within about 1 percent. However, if I^{131} were to be counted, the error would be as much as 3 or 4 percent assuming the same voltage regulation.

A Directional In-vivo Counter

An outline drawing of the directional *in-vivo* scintillation counter which has been used in this laboratory for the localization of Fe^{59} in human subjects is shown in Fig. 6. The phosphor is a one inch cubic piece of thallium activated sodium iodide located just behind the aperture and in contact with the phototube face. Anthracene and stilbene crystals can be used as phosphors but the counting efficiency for crystals of the same size is about one third as high because of the lower density and lower atomic number of these materials compared to sodium iodide. The plateaus obtained are about the same as the ones shown for the other counters using the same phosphor and a similar optical arrangement.

The counting efficiency when thallium activated sodium iodide is used is such that 38 percent of the gamma-rays from Co^{60} or Fe^{59} which go through the aperture are counted. The background is about 100 counts per minute. The efficiency is higher than a G-M counter by a factor of about 55, since the efficiency of a brass wall G-M counter to 1.2 Mev gamma-rays is approximately 0.7 percent.[10] The background of the scintillation counter is higher by a factor of about 5 assuming a normal background of about 20 counts per minute for a G-M gamma counter. Therefore, the sensitivity of the scintillation counter to weak sources is higher by a factor of about 11 over a G-M counter.

A curve showing the directionality of the counter when a 7/8 inch aperture is used is shown in Fig. 7. It shows that a sample gives half as strong a response when it is 15.8^0 off the axis of maximum counting. Greater or less directionality can be obtained, of course, by changing the size of the aperture. Two inches of lead shielding are used on all sides of the crystal and the phototube. This provides adequate shielding for isotopes having a gamma-ray energy slightly above 1 Mev or less.

A magnetic shield should be provided around the phototube to minimize the effects of the earth's magnetic field if it is desired to point the counter in different directions. Magnetic fields can change the gain of the phototube and therefore change the efficiency of the counter.

The electronic circuit of the counter is described in the next section. The entire phototube circuit including the cathode follower is located inside the lead shield in order to shorten the signal lead between the phototube and cathode follower. The cables leading to the scaler can then be several yards long if desired.

Circuit Details

A diagram of the phototube circuit is shown in Fig. 8. It is designed to connect directly to a scaling circuit ordinarily used for G-M counting.* In effect, the addition of this circuit converts the G-M scaler into a scintillation counter.

In order to make it possible to connect the phototube directly to the G-M scaler without an additional amplifier or power supply, a high load resistance is used in the anode lead of the phototube. Pulses are developed across this load resistance which are large enough in amplitude and duration to operate the scaler without additional amplification, providing the capacitance across the load resistance is low. The capacitance is kept as low as possible by using a cathode follower to feed the signal to the shielded cable that connects the phototube circuit to the scaler. The resulting pulses operate the scaler directly with less than the maximum rated voltage of 1250 volts applied to the phototube.

In previously reported experiments on scintillation counting, the phototube voltage is kept constant and a variable pulse height discriminator is used to investigate the various pulse heights produced. We have found it convenient for our purposes to set the scaler sensitivity to a fixed value, usually 0.1 volt or less, and then vary the high voltage applied to the phototube to change the sensitivity of the counter. The resulting curves are easy to interpret from a practical point of view since they are similar to the plateau curves obtained from G-M counters.

The operating voltage as read on the scaler voltmeter is 56 percent greater than the actual voltage applied to the phototube. The scaler we used contains a 1 megohm protective resistance in the high voltage lead which causes the voltage at the scaler high voltage terminal to drop when any appreciable current is drawn

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* We have used a Nuclear Instrument and Chemical Corporation Model 163 scaler.

through it. A small amount of current is drawn by the voltage divider to which the phototube dynodes are connected. The resistor was left in the scaling circuit partly as a safety precaution and also because it makes it slightly easier to maintain a constant voltage on the phototube.

An important requirement for stable operation which has not been mentioned yet is that the input sensitivity of the scaler must remain constant. This is a necessary condition to prevent changes in the counting efficiency since a scintillation counter is a proportional counting device. The scaler we have used is fairly satisfactory in this respect although it has been observed that over periods of several months, changes in input sensitivity have occurred which required slight readjustment of the scaler input sensitivity or the phototube operating voltage in order to maintain the same counting efficiency. Changes in sensitivity are of course detected by the regular counting of a standard sample. A scaling circuit designed to have an input sensitivity which is constant over long periods of time should prevent this trouble.

Acknowledgments

The author wishes to thank Dr. C. A. Tobias for his interest in this work, Joseph Mather for his participation in the early phases of the work, and Robert Mather and Robert Leininger for the preparation of some of the anthracene and stilbene crystals used. The equipment described here is presently being used by Dr. John H. Lawrence, Dr. Rex Huff, and other members of this laboratory in the study of Fe^{59} metabolism.

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Information Division
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9. Robert Hofstadter and John A. McIntyre, Nucleonics <u>7</u>, No. 3, 32 (1950).
10. R. F. Hill, G. J. Hine, and L. D. Marinelli, American Journal of Roentgenology and Radium Therapy <u>63</u>, No. 2, 160 (1950).

Figure Captions

Fig. 1 Anthracene flat crystal counter plateau

Fig. 2 Outline drawing of scintillation vial sample counter

Fig. 3 Thallium activated sodium iodide scintillation vial counter plateau

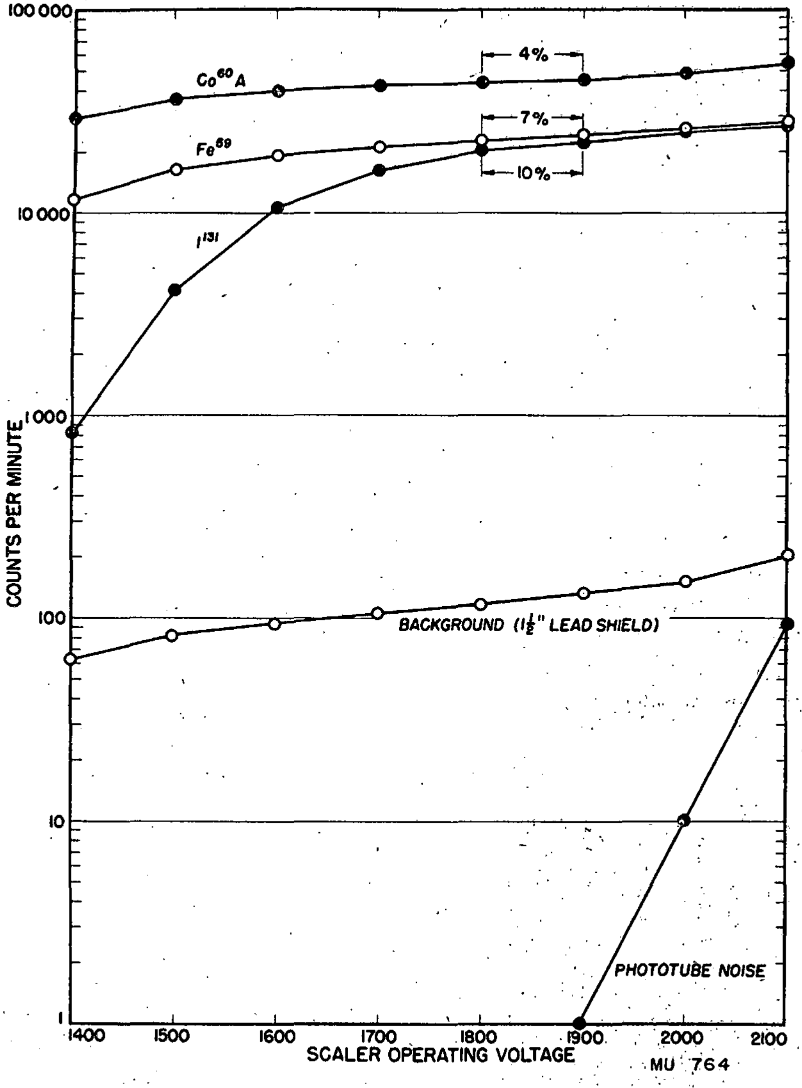
Fig. 4 Vial counter sample volume vs. Co^{60} counting efficiency

Fig. 5 Anthracene scintillation vial counter plateau

Fig. 6 Outline drawing of *in-vivo* scintillation counter

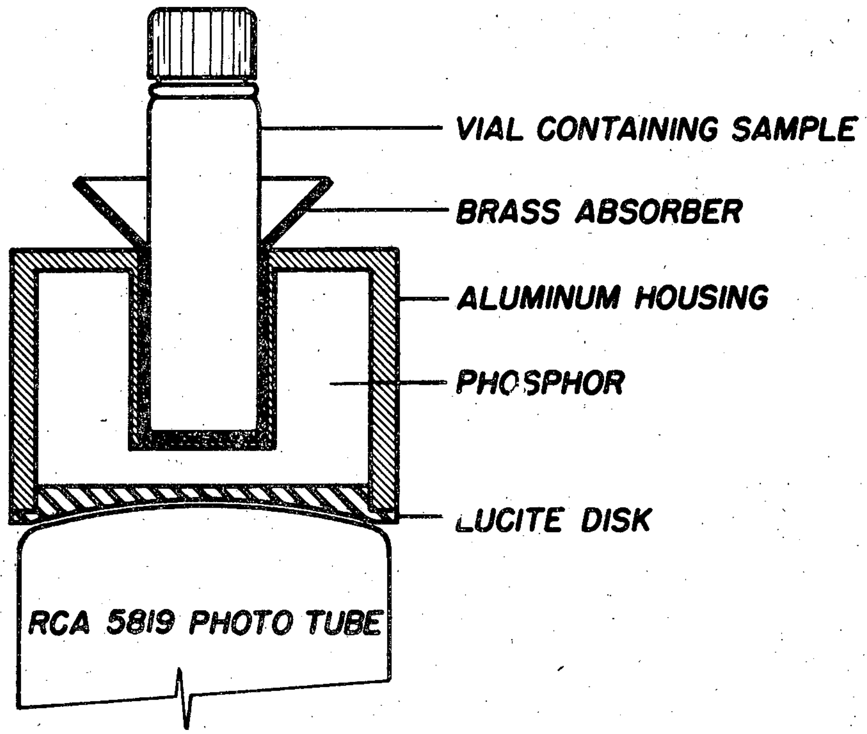
Fig. 7 Directionality of *in-vivo* scintillation counter

Fig. 8 Diagram of phototube circuit



100 000
10 000
1 000
100
10
1
COUNTS PER MINUTE
1400
1500
1600
1700
1800
1900
2000
2100
SCALER OPERATING VOLTAGE
Co^{60}A
4%
7%
10%
Fe^{59}
I^{131}
BACKGROUND (1½" LEAD SHIELD)
PHOTOTUBE NOISE
MU 764

Fig. 1


VIAL CONTAINING SAMPLE
BRASS ABSORBER
ALUMINUM HOUSING
PHOSPHOR
LUCITE DISK
RCA 5819 PHOTO TUBE

MU 1327

Fig. 2

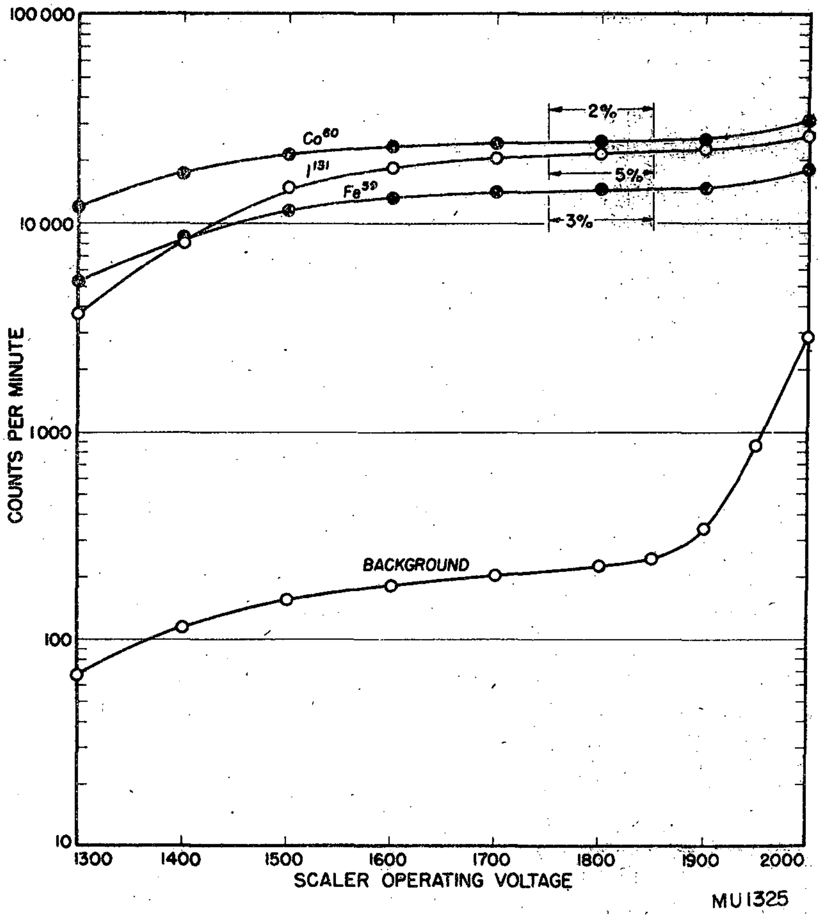

100 000
10 000
1000
100
10
COUNTS PER MINUTE
1300
1400
1500
1600
1700
1800
1900
2000
SCALER OPERATING VOLTAGE
Co^{60}
I^{131}
Fe^{59}
BACKGROUND
2%
5%
3%
MU 1325

Fig. 3

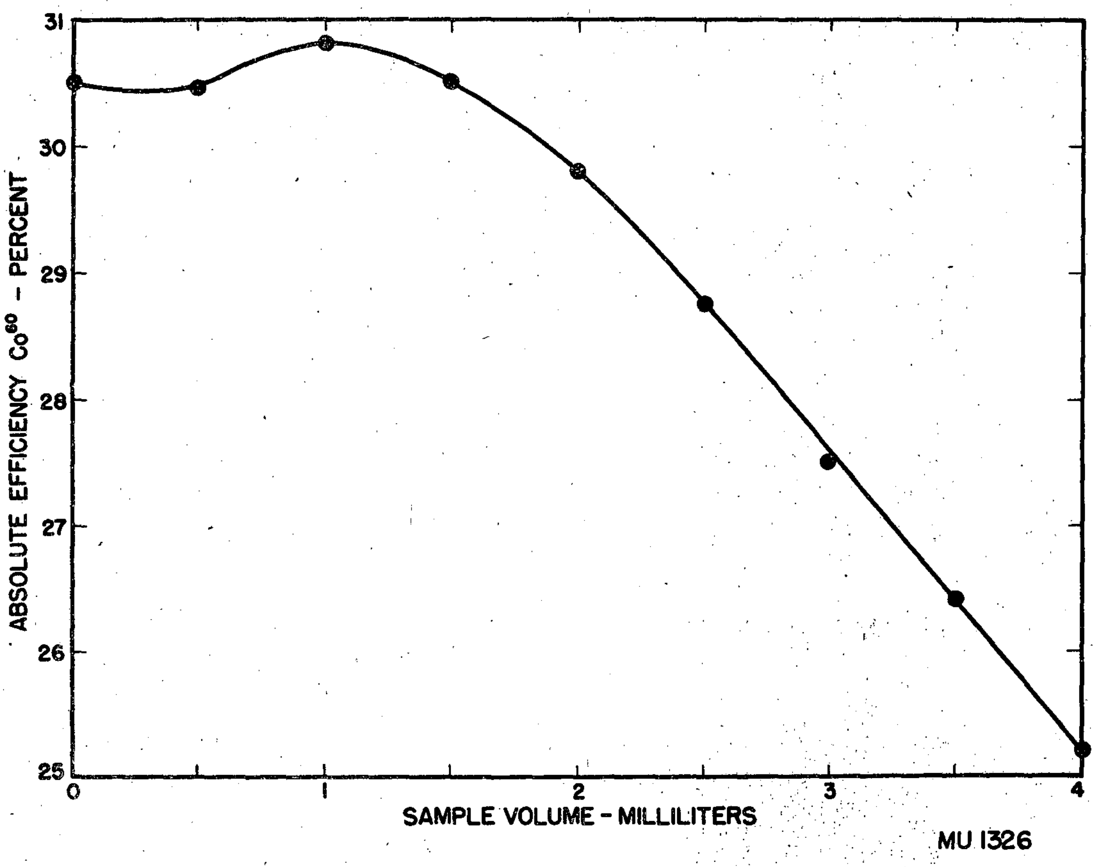
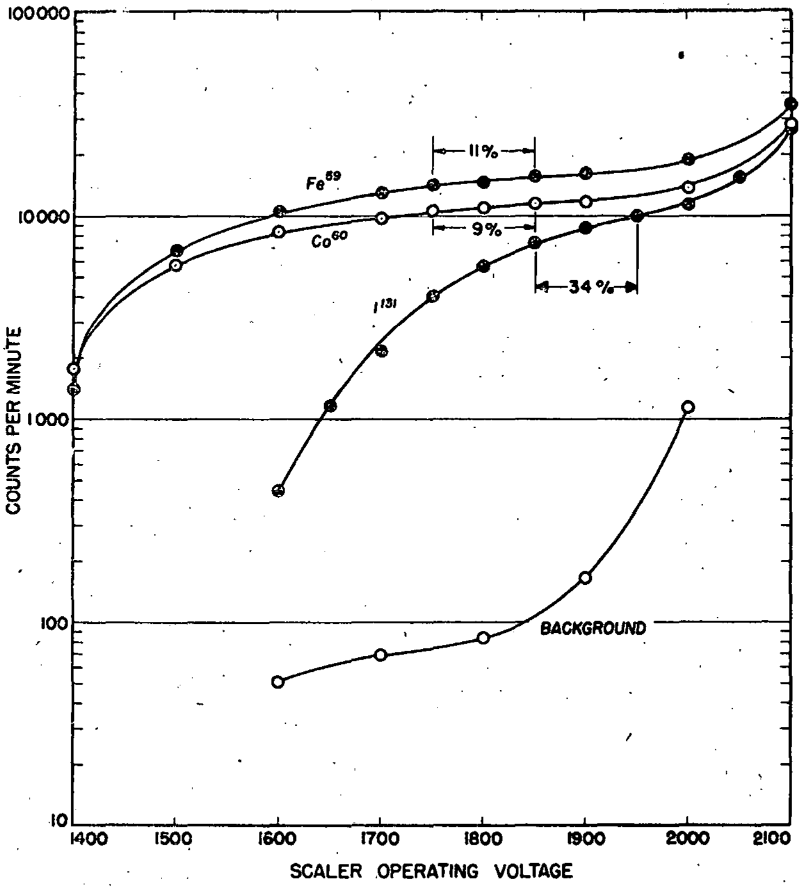

31
30
29
28
27
26
25
ABSOLUTE EFFICIENCY Co^{60} - PERCENT
0
1
2
3
4
SAMPLE VOLUME - MILLILITERS
MU 1326

Fig. 4


100000
10000
1000
100
10
COUNTS PER MINUTE
1400
1500
1600
1700
1800
1900
2000
2100
SCALER OPERATING VOLTAGE
Fe^{59}
Co^{60}
I^{131}
BACKGROUND
11%
9%
34%

MU 763

Fig. 5

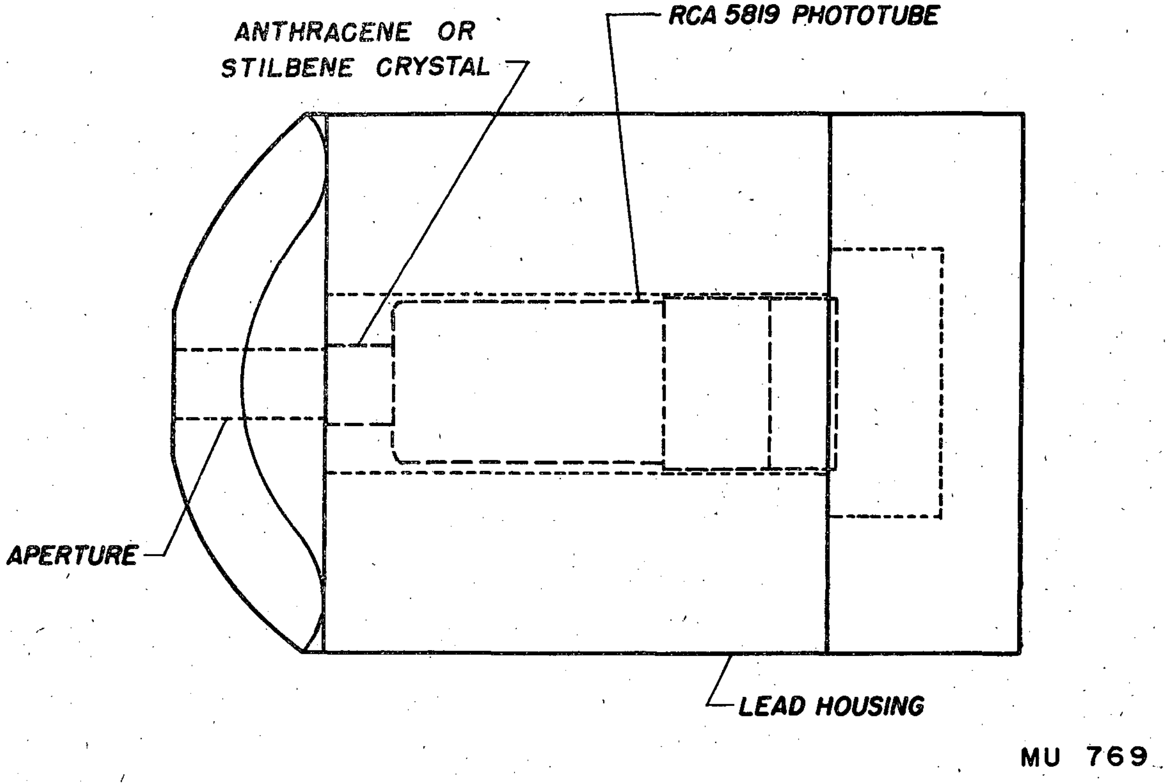

ANTHRACENE OR STILBENE CRYSTAL
RCA 5819 PHOTOTUBE
APERTURE
LEAD HOUSING
MU 769

Fig. 6

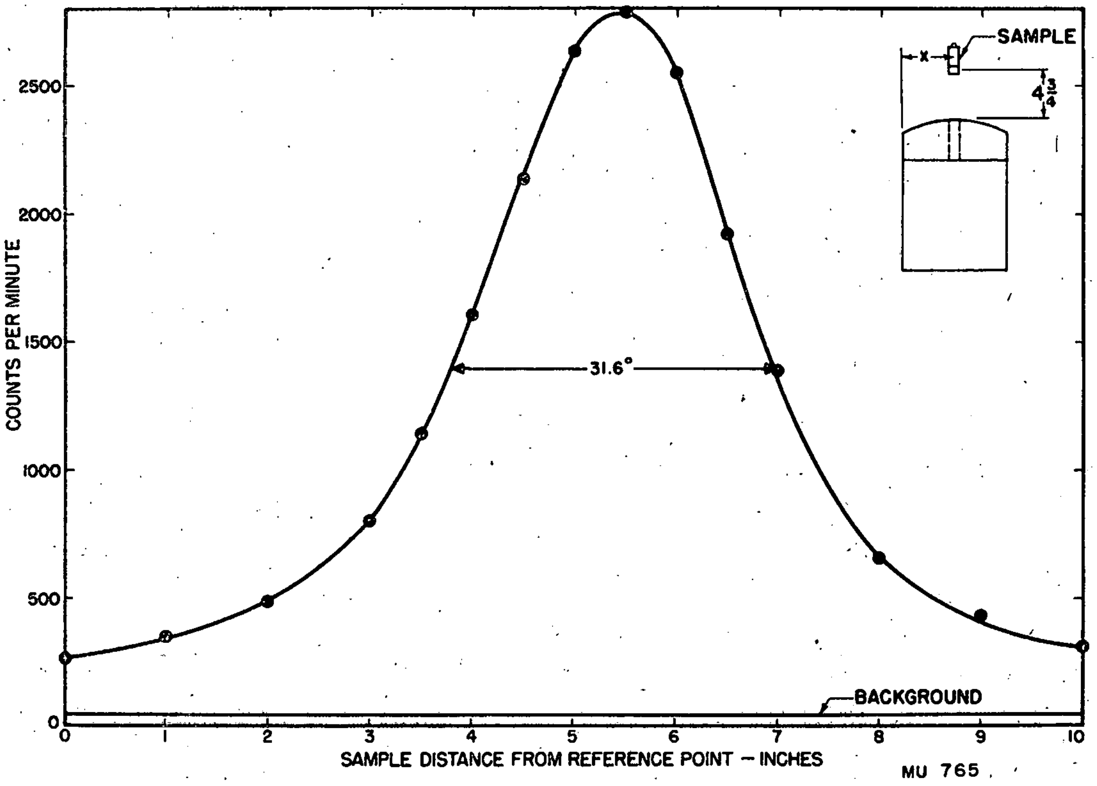

SAMPLE
X
4 3/4
31.6°
BACKGROUND
COUNTS PER MINUTE
0
500
1000
1500
2000
2500
0
1
2
3
4
5
6
7
8
9
10
SAMPLE DISTANCE FROM REFERENCE POINT — INCHES
MU 765

Fig. 7

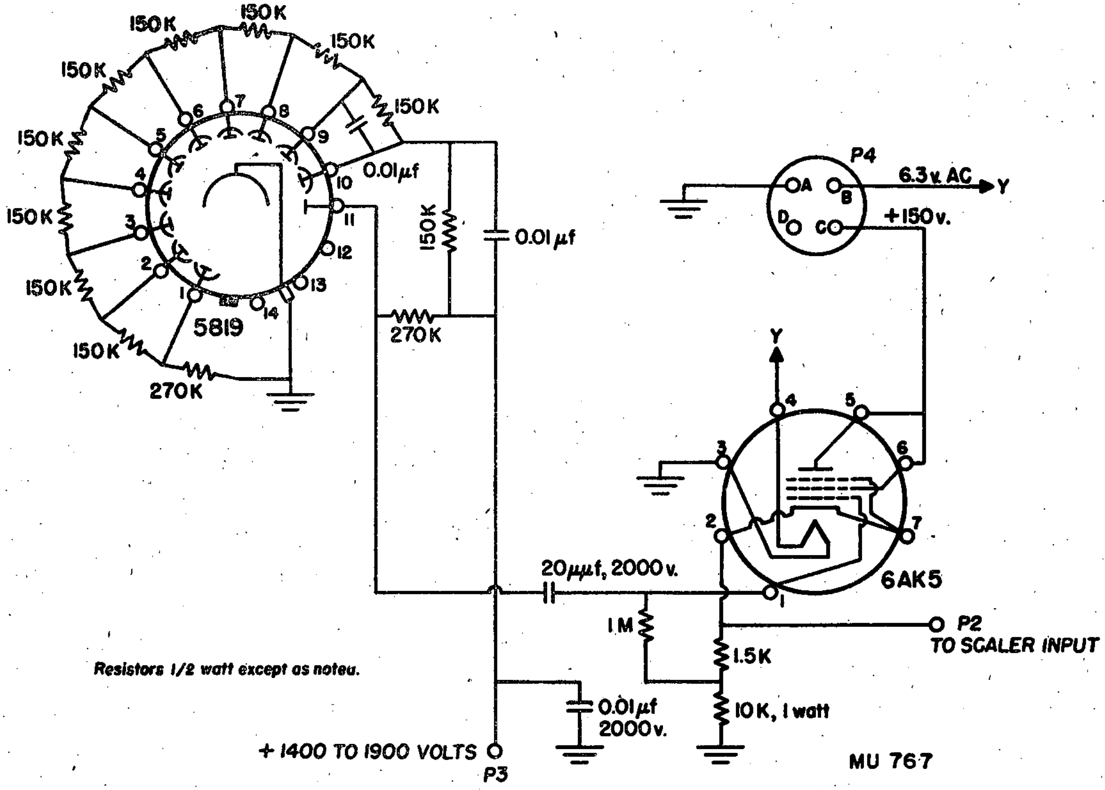

150K
150K
150K
150K
150K
150K
150K
150K
150K
270K
5819
1
2
3
4
5
6
7
8
9
10
11
12
13
14
0.01μf
150K
270K
0.01μf
P4
A
B
D
C
6.3v AC
Y
+150v.
Y
4
5
3
6
2
7
1
6AK5
20μμf, 2000v.
1M
1.5K
P2
TO SCALER INPUT
Resistors 1/2 watt except as noted.
0.01μf
2000v.
10K, 1 watt
+ 1400 TO 1900 VOLTS
P3
MU 767

Fig. 8