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SCINTILLATION COUNTERS FOR THE MEASUREMENT OF RADIOACTIVE SAMPLES

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Scintillation Counters for the Measurement of Radioactive Samples

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University of California, Berkeley, California

August 30, 1950

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 avaiable. In this laboratory we have attempted to apply scintillation counting to some of our problems in biological tracer research.

The great advantage of scintillation counting is that it makes possible the detection of gamma-radiation with much greater efficiency than is possible with Geiger counters. This opened the possibility that we might be able to count gamma-rays from some of our samples instead of beta-particles as we have been doing previously. Samples containing Fe^{59} and Co^{60} are examples. Both of these isotopes emit beta-particles of relatively low energy, Co^{60} having a maximum energy of 0.3 Mev, and Fe^{59} having two beta-particles with energies of 0.26 and 0.46 Mev. 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 reproducability because of the involved chemical procedure through which the

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[1] H. Kallman, Natur und Tecknick, July 1947.

[2] M. Deutsch, Massachusetts Institute of Technology Technical Report No. 3 (December 1947).

[3] Harshaw Chemical Co., Cleveland 6, Ohio.

biological samples had to be put in order to prepare them for counting. This procedure consisted of electroplating the samples on metal discs in order to get samples that were thin enough so that the self absorption of the samples would be small. Counting gamma-rays with a scintillation counter makes it possible to eliminate the electroplating and all the chemical procedure leading up to it. The activity of the samples in most cases can be measured merely by placing them in glass vials without any other preparation and then placing the vials in the counter.

The gamma-ray counting efficiency with the scintillation counter is not quite as high as for beta counting with a G-M counter. When counting Co^{60} it is about three-fourths as efficient, and when counting Fe^{59} the factor is one-half. The background count in each case is about three times as high for scintillation counting. In spite of this, however, this method has such great advantages over the older method of sample measurement that it is preferred for work with Co^{60} and Fe^{59}, and will no doubt prove useful with many other gamma emitting isotopes.

Principles of Scintillation Counting

An important requirement of a counter for radioactive samples is that it must be adequately stable. It was at first thought that this might be a difficult requirement to meet since a stable counter would be very difficult to construct if a counting plateau of some kind did not exist. We have found that a reasonably satisfactory plateau is obtained for gamma-rays with energies of about 1 Mev and greater when a suitable phosphor and a selected 5819 phototube are used at room temperature. Anthracene and stilbene are satisfactory phosphors. 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, we find that 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}.*

The I^{131} plateau is observed to have a steeper slope than the Fe^{59} plateau, and furthermore, 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

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[4] W. V. Mayneord, E. H. Belcher, Nature, 165, 930 (June 10, 1950).

* 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.

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.

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 practically all the scintillations of a certain magnitude produced in the crystal are being counted. The fact that the count drops practically to zero when the crystal is removed indicates that phototube noise is not being 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. It is apparent that 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 two phosphor materials mentioned above. It is likewise apparent that such measures will be necessary for counting very low energy gamma-rays. Other experimenters have reported on these methods.[2,5,6]

The resolving time or insensitive time of one of the above counters was 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.[7] The overall resolving time of the counter is adequate for all the usual counting requirements without correction for resolving time since the correction is only 0.1 percent for every 10,000 counts per minute.

The Vial Sample Counter

As mentioned before, we have found it advantageous to count samples which are contained in glass vials. A drawing showing a counter for these samples is given in Fig. 2. The crystals are arranged around a well in which the sample vials are placed. Thus the counting geometry is fairly high. 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-rays from entering the crystal. The brass absorber which fits between the vial and the aluminum housing prevents the housing from becoming contaminated by activity that may be picked up from contact with the sample vials, and also provides part of the absorption path to prevent beta-rays from entering the phosphor from the sample. If it becomes contaminated from contact with the samples, it can easily be removed and cleaned.

A plateau curve is shown in Fig. 3 for a counter of this type which uses anthracene crystals. It will be noted that the Fe^{59} curve has a slope of

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[5] G. A. Morton, K. W. Robinson, Nucleonics <u>4</u>, No. 2, 25 (1949).

[6] R. F. Taschek, H. T. Gittings, Phys. Rev. <u>75</u>, 1553 L (1949).

[7] George Kelley, Oak Ridge National Laboratory Report No. 366 (June 1949).

about 11 percent per hundred volts and the Co^{60} curve has a slope of about 9 percent per hundred volts when operating at 1800 volts. The I^{131} curve has a slope of 34 percent per hundred volts when operating at 1900 volts. The increased slope of the plateau of this counter is probably due to the decreased efficiency of light transmission between the crystals and phototube. While the slopes for Fe^{59} and Co^{60} are not as low as for a good G-M counter, they are still satisfactory since it is possible to control the high voltage from a good scaler to within 10 volts, and any error due to variation in the high voltage is therefore kept to about 1 percent. If I^{131} were counted, the error would be as much as 3 percent or 4 percent assuming the same voltage regulation. This type of scintillation counter is not as practical for the lower energy gamma emitters.

The absolute counting efficiency of a typical counter of this type which accepts 1 dram glass vials and uses anthracene crystals is 7.0 percent for Fe^{59} and 13.1 percent for Co^{60} when the vials are filled with 2 milliliters of solution. The efficiency is 6.4 percent for Fe^{59} and 11.9 percent for Co^{60} when the vials are filled almost full with 4 milliliters of solution. The absolute counting efficiency is defined as the counts per minute recorded by the counter divided by the disintegration rate of the sample in disintegrations per minute.

The background at the most satisfactory operating voltage is about 80 counts per minute when the counter is shielded with 4 inches of lead plus 2 inches of iron next to the counter, and about 120 counts per minute when shielded with 1 1/2 inches of lead plus 1/4 inch of aluminum next to the counter. The background when the counter is unshielded is about 400 counts per minute. Examination of Fig. 3 shows that the background can be varied a great deal by varying the operating voltage of the scaler. Since the counting efficiency changes at the same time, a compromise is necessary to obtain the best condition for counting weak samples.

Fig. 4 shows a curve of the efficiency of this counter versus the volume of the sample contained in the vial. This curve is of use when counting liquid samples of various volumes. It will be noted that the efficiency remains approximately constant until the volume reaches 2.5 milliliters, after which it begins to decrease slightly. Samples which occupy 2.5 milliliters or less can be counted at a known absolute efficiency without knowing the exact volume of the sample. Thus it is possible to count small bits of biological tissue by merely packing them into the bottom of the vial and counting them in the regular manner.

When the absolute counting efficiency for Co^{60} is known for a gamma counter, the absolute efficiency for Fe^{59} can be calculated from the following formula:

$$E_{Fe} = \frac{E_{Co}}{2 - E_{Co}}$$

Where E_{Fe} = Absolute counting efficiency for Fe^{59}

E_{Co} = Absolute counting efficiency for Co^{60}

This formula is derived from the fact that the two isotopes emit gammas of almost exactly the same energy, the difference being that each Co^{60} disintegration yields two gammas in coincidence with energies of 1.1 and 1.3 Mev, while each Fe^{59} disintegration yields one gamma with a 50 percent chance of its being either the 1.1 or 1.3 Mev energy. The Co^{60} efficiency, therefore, is slightly less than twice the Fe^{59} efficiency, the slight loss being due to coincident counts in the case of Co^{60}.

Circuit Details

A diagram of the phototube circuit is shown in Fig. 5. It is designed to connect directly to a scaling circuit ordinarily used for G-M counting.* In effect, the addition of the phototube circuit converts the G-M scaler into a scintillation counter.

Two minor changes have been made in the phototube circuit as it is usually used to make it possible to use the G-M scaler without an additional amplifier or power supply. First, a high load resistance, 270,000 ohms, 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 with no additional amplification, providing the capacitance across the load resistance is low. The capacitance is kept as low as possible by using a 6AK5 tube as a cathode follower to feed the signal to the shielded cable that connects the phototube circuit to the scaler. The resulting output operates the scaler directly with less than the maximum rated voltage of 1250 volts applied to the phototube.

No variable pulse height discriminator is used. Instead the scaler sensitivity is set at a fixed value, usually at 0.25 volts, and the high voltage applied to the phototube is varied to change the sensitivity of the counter. 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 through it. A small amount of current is drawn by the voltage divider to which the phototube dynodes are connected.

As mentioned before the voltage supplied to the phototube is not unduly critical as long as a phosphor is used which gives a moderately good counting plateau. Another requirement for stable operation which has not been

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

mentioned is that the input sensitivity of the scaler must remain constant. The scaler we have used is fairly satisfactory in this respect although it has been observed that changing the two input tubes of the scaler results in a different input sensitivity, requiring readjustment of the scaler input sensitivity or the phototube voltage to maintain the same counting efficiency.

A Directional In-vivo Counter

An outline drawing of the directional *in-vivo* scintillation counter which has been used in this laboratory is shown in Fig. 6. The crystal is a one-inch cubic piece of stilbene located just behind the aperture and cemented to the phototube face. Two inches of lead shielding are used on all sides of the crystal and phototube. This provides adequate shielding for isotopes having a gamma-ray energy slightly above 1 Mev, such as Fe^{59} and Co^{60}.

The efficiency is such that about 10 percent of the gamma-rays which go through the aperture are counted. The background is about 40 counts per minute. A curve showing the directionality of the counter is shown in Fig. 7. It shows that a sample gives half as strong a response when it is 15.8° off the axis of maximum counting. Greater or less directionality can be obtained, of course, by changing the size of the aperture.

The electronic circuit is the same as for the counters just described. The entire phototube circuit including the cathode follower is located inside the lead shield in order to shorten the signal lead between phototube and cathode follower. The cables leading to the scaler can be several yards long if desired.

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. The shield can be made of a piece of cold rolled steel tubing with about a one-fourth inch wall thickness, and it should extend the full length of the phototube.

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 Lawrence, Dr. Rex Huff, and other members of this laboratory to study Fe^{59} metabolism. It is expected that information will be published about its use in this connection.

This work was performed under the auspices of the Atomic Energy Commission.

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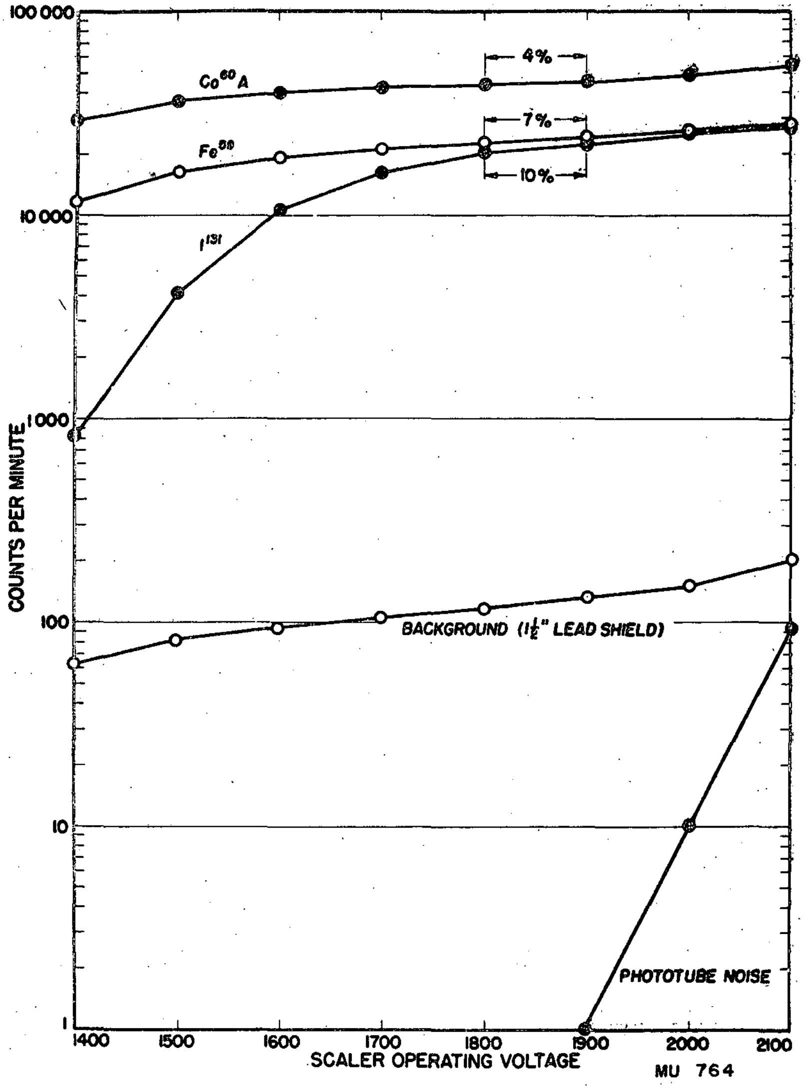

Fig. 1. Anthracene flat sample counter plateau.

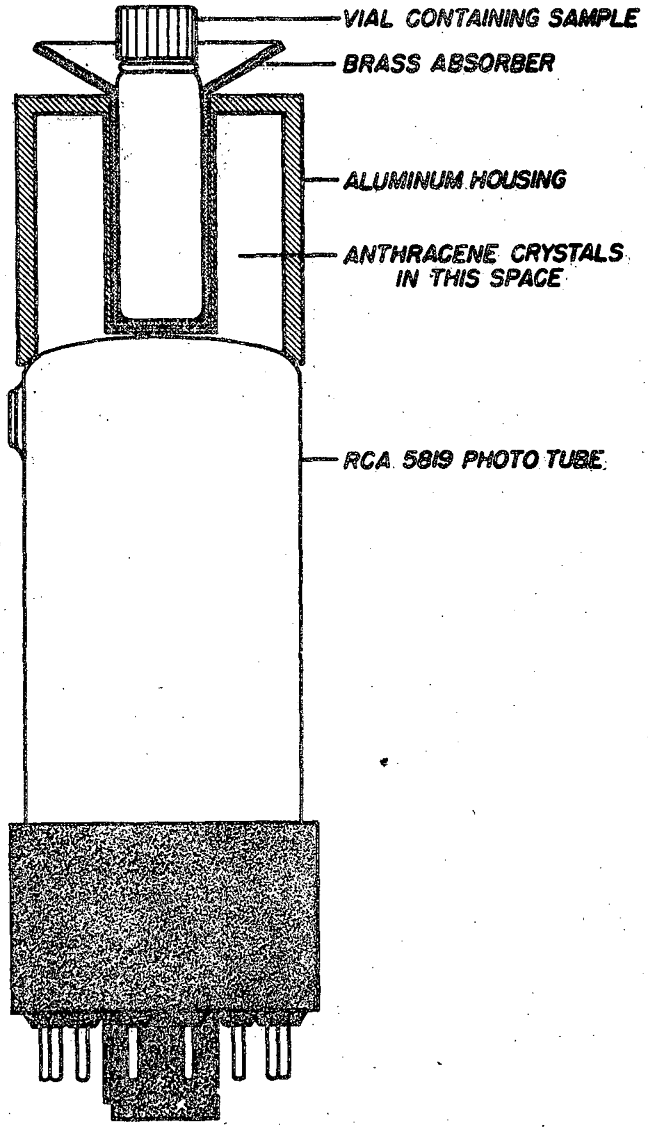

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Fig. 2. Outline drawing of scintillation vial sample **counter.**

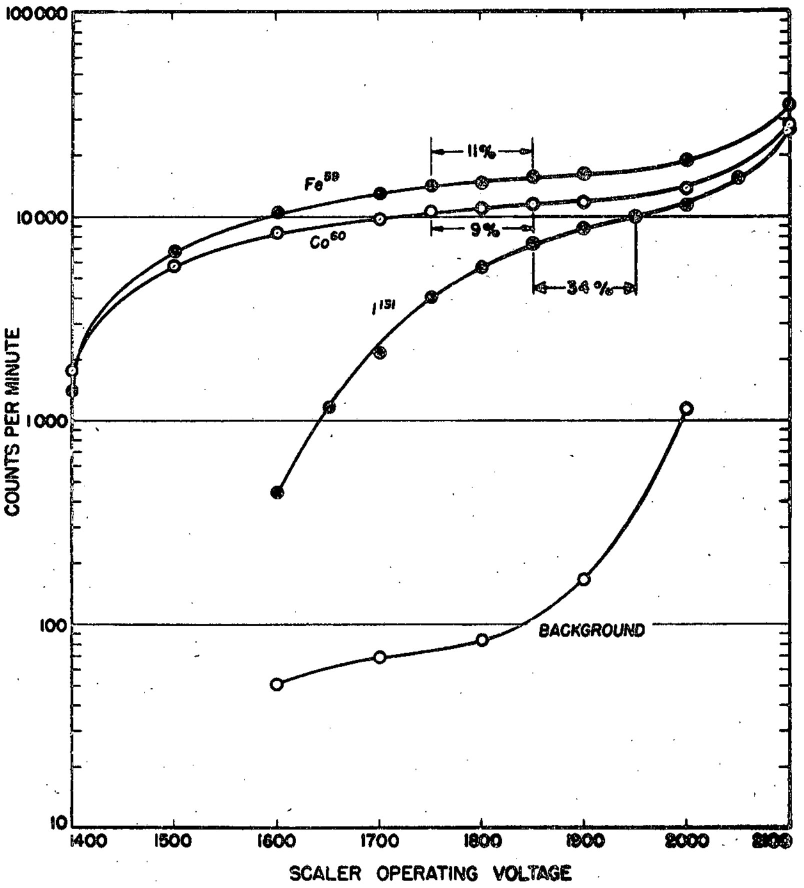

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Fig. 3. Scintillation vial **counter plateau.**

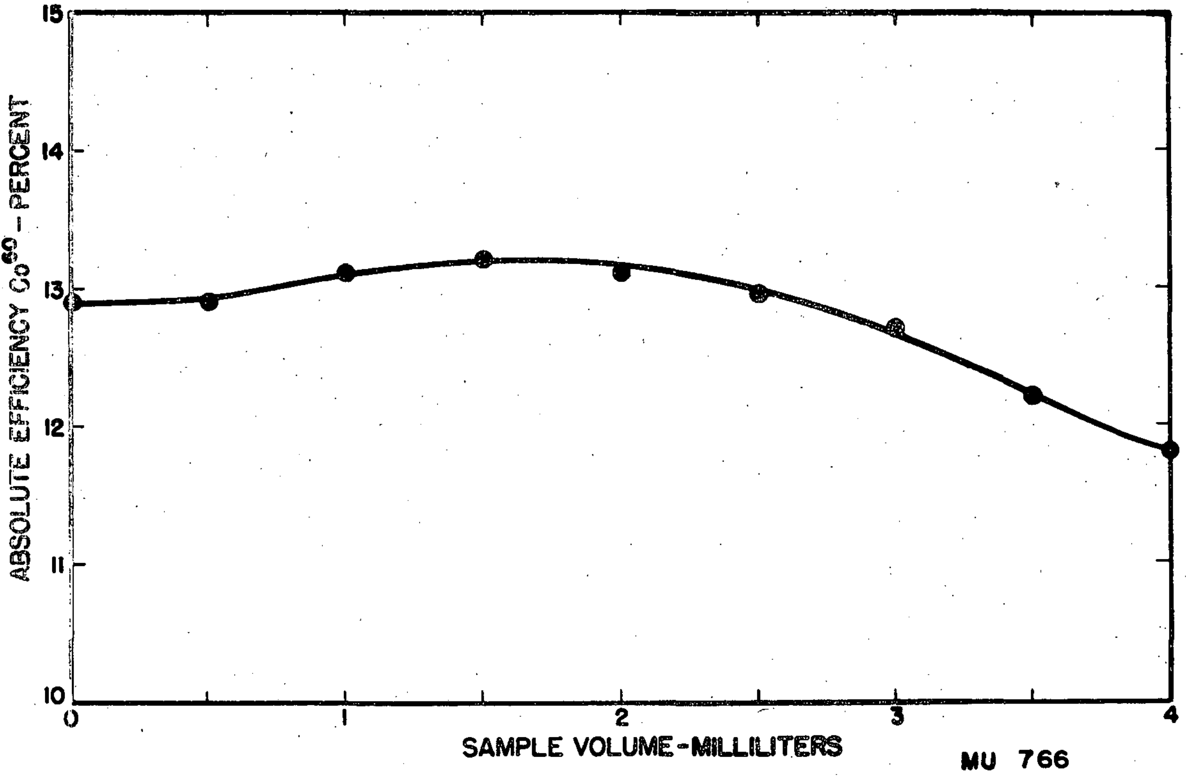

Fig. 4. Scintillation vial counter, sample volume vs. Co^{60} efficiency.

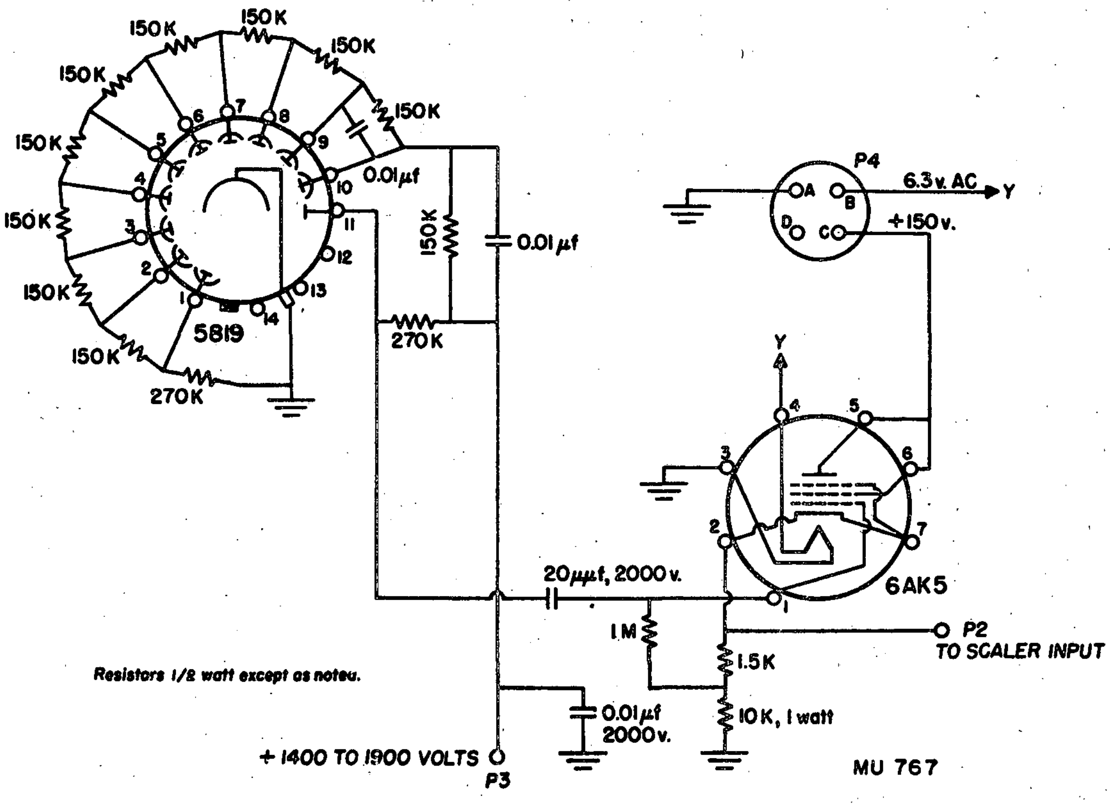

Fig. 5. Wiring diagram of scintillation counter.

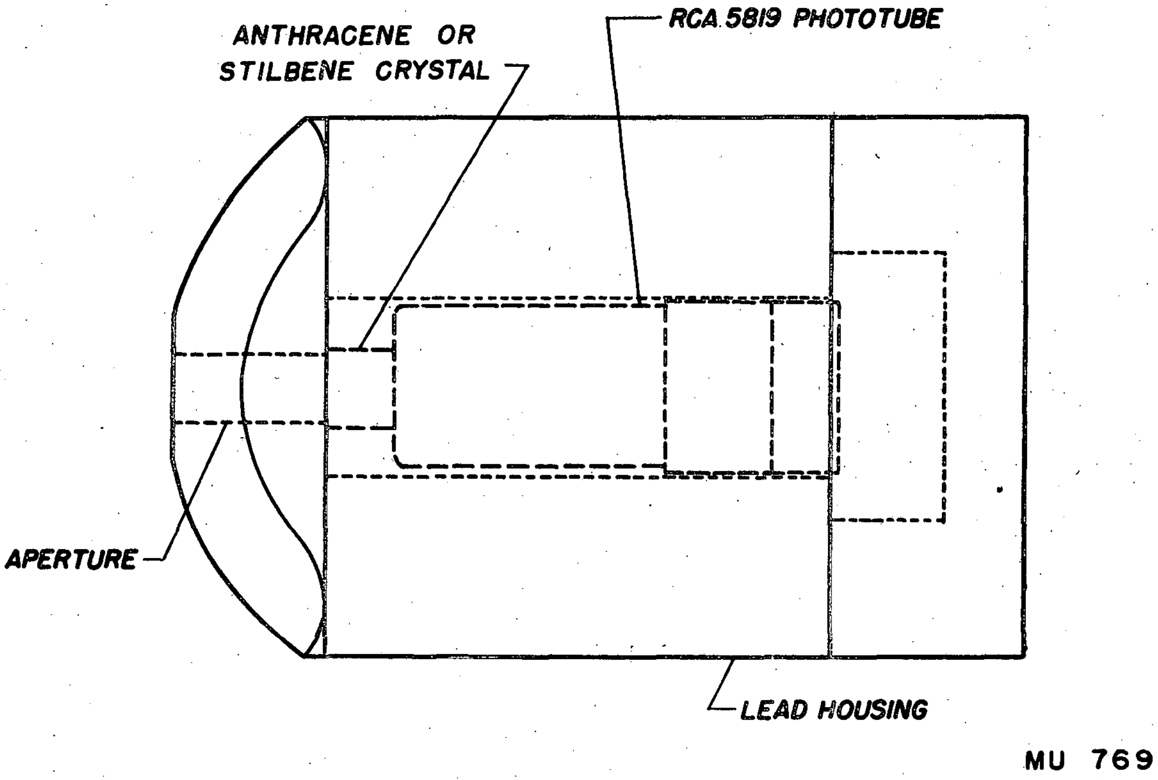

Fig. 6. Outline drawing of in-vivo scintillation counter.

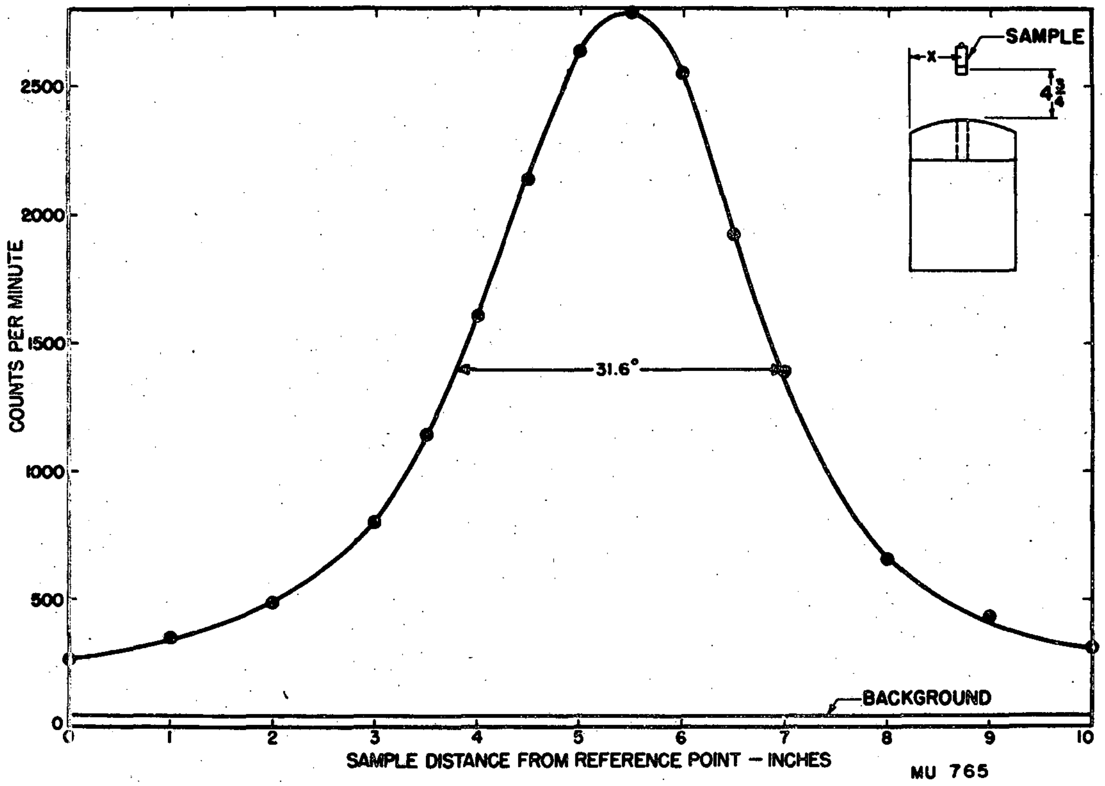

Fig. 7. Directionality of in-vivo gamma counter.