

# THE DESIGN OF A COMMERCIAL LIQUID SCINTILLATION COINCIDENCE COUNTER

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THE liquid scintillation coincidence counter is an instrument which receives weak light signals from a scintillating solution and translates them into a record of counted events. The first phase of this detection process uses a device which both produces electrons in exchange for light quanta and which multiplies the number of these electrons by many factors of ten. This unit, the photomultiplier, has the disadvantage of yielding electrons without incident light. Such background is very high under conditions wherein weak beta-emitters would be counted. The coincidence feature, in conjunction with refrigeration, reduces this background without interfering with desired signals. The instrument, therefore, has the photomultipliers viewing the counting bottle, all of which is placed in a refrigerated chamber. Shielding of the scintillator solution from external background is effectively accomplished by surrounding the whole photomultiplier-cell assembly with a 2 in. thickness of lead. The scintillation light which is not emitted toward a photomultiplier reaches a very efficient reflector whose purpose is to redirect photons until they eventually reach the photosensitive surface and become converted into an electrical pulse.

The first complete practical liquid scintillation coincidence counter was made by HIEBERT, HAYES and WATTS of the Los Alamos Scientific Laboratory. This instrument was a great advance forward and enabled the counting of carbon-14 with efficiencies of 60-70% and the counting of tritium with efficiencies of from 10 to 15%. This instrument, after which a commercial instrument was originally patterned, utilized two fast linear amplifiers, fast coincidence to cut down photomultiplier tube noise and simple anticoincidence to provide a 'window', so that high amplitude noise pulses were rejected. One scaler was used, so that the instrument was capable of counting one isotope at a time.

I would like to describe briefly the path of attack which we took in designing a liquid scintillation coincidence counter. Our objectives were to produce a liquid scintillation counter which could be used in routine laboratory procedures by inexperienced personnel. We desired a maximum carbon-14 efficiency of 80% or better and a maximum tritium efficiency of 15-20%. Naturally, we wished the lowest possible background count for the completed

instrument. Thus, we set the maximum permissible count rate from accidentals at 5 counts/min at a temperature of 0°C. We decided that the instrument should be capable of counting two isotopes simultaneously in a given sample. For this reason, we set the dynamic discriminator range specification to be 20-1 or better. In designing our liquid scintillation coincidence counter, it was decided to eliminate or circumvent a number of difficulties then present in available liquid scintillation counters. Some of these difficulties were as follows:

1. Increase of background rate, due to removing high voltage when sample was changed.
2. Varying efficiency, due to the use of oil coupling. The use of oil optical coupling, while resulting in increased inefficiency over air coupling, produced two undesirable effects. First, with continued use, the oil became contaminated. While this was not visually noticeable, the contamination did absorb ultraviolet radiation so that the counting efficiency decreased with time as more and more samples were counted. Second, the oil coupling increased the background, due to the production of Čerenkov radiation.
3. Amplifier overloading, when counting of two isotopes simultaneously, especially isotopes that differ greatly in energy. Severe demands were made on the linear pulse amplifiers currently used, and they could not operate successfully under these conditions.

The exact design of our liquid scintillation coincidence counter will now be described. Each component in the system, starting with the sample holder, will be discussed.

The sample holder was designed to hold either a vial containing from 5 to 25 ml of scintillator or a bottle containing from 10 to 70 ml of scintillator. An adapter was used for the smaller vial, so that it was held rigidly in position and so that light loss was decreased. It was found that polished aluminum was rather a flat reflector with respect to wavelength, and thus it was insensitive to the addition of POPOP wavelength shifter. Samples without the wavelength shifter would count with approximately the same efficiency with a polished aluminum reflector or a flat white reflector, such as white Tygon paint. However, it was found that the combination of white Tygon and samples using wavelength shifter showed marked improvement in efficiency. In spite of the increased efficiency, it was decided to use a polished aluminum reflector because of its much greater durability.

The sample holder was designed with a cylindrical shutter, so that when the sample was loaded into the sample holder, light could not reach the photomultiplier tubes. In order to positively prevent the top of the sample holder from being removed without the shutters being closed, a mechanical interlocking feature was used, so that it was impossible to remove the sample holder cap without rotating the shutter to a closed position. By using this

interlocking feature, it was possible to change samples without removing the high voltage from the photomultiplier tubes. Thus, there was no change in sensitivity or any increase in background produced by this sample changing operation.

The sample holder assembly utilized spring loaded photomultiplier tubes and mu-metal magnetic shields. The whole assembly was designed as a light-tight unit, so that it could easily be dropped in the 2 in. thick lead shield without exposing the photomultiplier tubes. Since the sample holder-photomultiplier tube assembly was light-tight, the freezer cover could be left open while routinely counting a large number of samples. The freezer cover was not required to be light-tight.

There are at least two methods of logic which may be used in coincidence pulse height analyzers suitable for liquid scintillation counting. The original Los Alamos counter and the other available instrument patterned after it used a system of logic wherein one multiplier tube operates at near maximum gain at all times and is used as a noise detection channel. This channel has only a lower level discriminator and its output is put into coincidence with the other channel, which is used for pulse height analysis. This unsymmetrical system results in more coincidence noise under all conditions of operation except when the instrument is operated at absolute maximum gain. Also, since the output of a fixed low level discriminator is put in coincidence with another discriminator operated at a different sensitivity, the triggering points of the two discriminators differ in time because the amplifiers have a finite rise time. If a symmetrical system is used, wherein the voltages on the two photomultiplier tubes are essentially the same and the gains of the two amplifiers are identical, one can obtain minimum noise at any particular high voltage setting. In the system which we chose, the two lower level discriminators were put in coincidence, the two middle level discriminators were put in coincidence, etc. Therefore, if the two input signals had the same rise time and approximately the same amplitude, the triggering of any pair of discriminators would occur at the same point in time. Thus, with the amplifiers operating at a rise time of  $0.15 \mu\text{sec}$ , it is possible to obtain coincidence resolving times much shorter than this. The major advantage of using a symmetrical system is that for a given accidental noise rate due to random coincidences, the photomultiplier tube selection is greatly reduced.

The electronic circuits were arranged as shown in Fig. 1. Fast non-overloading amplifiers were used patterned after those developed by Chase-Higinbotham. Each photomultiplier-amplifier channel was fed into three discriminators, lower level, middle level and upper level, respectively. The two lower level discriminators are fed into a lower level coincidence tube to produce a low level coincidence pulse, as are the middle level discriminator pair and the upper level discriminator pair. Each Schmitt discriminator contains a critically damped inductance in the output plate, so that the

discriminator produces pulses approximately  $10^{-7}$  sec long, which are fed to the input of the coincidence circuits.

Thus far, we have followed the operation of the coincidence pulse height analyzer to the outputs of the lower, middle and upper level coincidence tubes. In order that one scaler (scaler *A*) represent all pulses lying above the lower

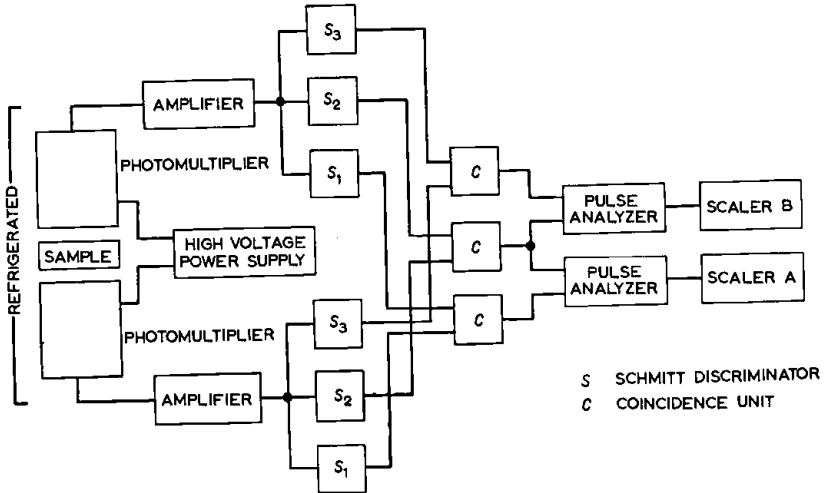


Fig. 1. Simplified block diagram—Model S-2A dual channel liquid phosphor counting system.

level but below the middle level, and scaler *B* represent all pulses lying above the middle level discriminator but below the upper level discriminator, it is necessary to impose a condition wherein, if a pulse appears at either of the respective upper levels, no pulse appears on the output. Instead of using a simple anticoincidence circuit, such as has been used on all other liquid scintillation coincidence counters, it was decided to use a more complex anticoincidence circuit, which would not be dependent upon pulse rise time or duration. Simple anticoincidence circuits work properly only when the two input pulses are alike in shape and duration, while the more complex circuit stores energy in a capacitor when the upper level has been crossed and puts this information in anticoincidence with the lower level information after both discriminators have been fired. In our system, the interrogate signal is produced by the recovery of the lower level discriminator. The coincidence pulse height analyzer operates well over a very large range of input pulse durations and amplitudes. It should be noted that using the system previously described, no pulses are lost if the input amplitudes of two coincidence signals are not identical. If, for instance, one phototube signal fired both the lower and middle level discriminators, while the other phototube signal fired only the lower discriminator, the output pulse would be counted in scaler *A* since only the lower coincidence tube produced an output.

The Schmitt discriminators used in the analyzer may be operated with discrimination levels of from 4 to 100 V, thus producing a dynamic range of 25-1. If two isotopes with widely different energies are to be counted, there is a provision for setting the middle discriminator to infinity so that all pulse exceeding the middle level will be counted in scaler *B*. Furthermore, it is possible to operate in a separated discriminator mode, so that one scaler counts all pulses lying between the lower level and middle level discriminator settings, while the other scaler counts all pulses lying above the upper level discriminator. This last method of operation has many advantages when two isotopes are counted simultaneously and their energy difference is quite low (3 : 1).

The two scalers are each composed of three decade scaling strips with 2.5  $\mu$ sec resolving time, followed by an electrically reset four-digit mechanical counter. Each decade strip utilizes etch-wired construction and is easily replaceable. The over-all resolving time for the entire system is better than 5  $\mu$ sec and an elapsed preset timer controls the count cycle to an accuracy of 0.2 sec.

Rather than have any amplifier gain controls to adjust the system for the counting of various isotope energies, a wide range high voltage power supply with excellent long time stability and regulation was used. Thus, the over-all system gain is controlled by a decade high voltage control, which selects voltage by hundreds, tens and units. The stability of the high voltage is approximately 200 p.p.m. per day, and any voltage may be selected and then reset to within a fraction of a volt.

The photomultiplier tubes are carefully selected by screening them for approximate equivalence in voltage gain. We did this by using a single phototube and a carbon-14 sample. The high voltage was increased until the carbon-14 just began to count at a particular discriminator setting. This value of high voltage is a measure of the photomultiplier tube gain. Tubes for a given instrument are selected, so that for equal gains the values of high voltage which must be applied to them do not differ by more than 200 V. The second measurement by which we selected photomultiplier tubes was based on their noise properties. Here again, we used a single phototube amplifier and discriminator and adjusted the value of high voltage applied to the photomultiplier to a point at which a standard tritium sample would count with a specified efficiency. We then removed the tritium sample and checked the noise rate for that particular photomultiplier. The preceding measurements were carried on at two different temperatures.

Our instrument might have been designed to count a single isotope rather than two isotopes simultaneously by pulse height selection. This would have eliminated two Schmitt discriminators, and anticoincidence circuit and one scaler. This simplification really changes the over-all complexity of the whole instrument by a very small factor, because a major portion of the cost of the

instrument is associated with the selection of the photomultipliers, testing, and with the linear amplifiers and power supplies. The additional economic gain is a small one and the use of two scalers carries out the over-all philosophy

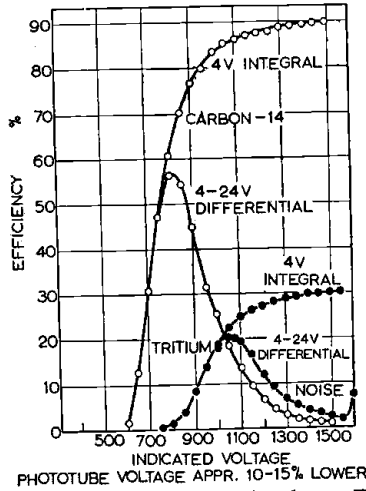


Fig. 2. Absolute counting efficiency vs. indicated voltage. Ten milliliter sample, plain aluminum reflector. Mk II sample holder with vial adapter.

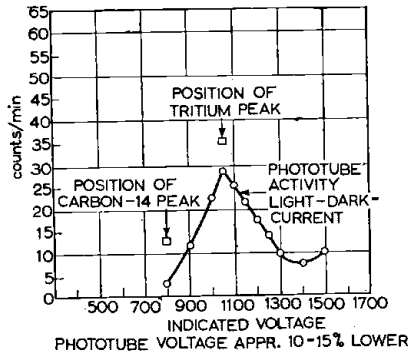


Fig. 3. Background vs. indicated voltage. Plain aluminum reflector. Mk II sample holder with vial adapter, temperature  $-10^{\circ}\text{C}$ , slit width 4.0-24 V, total background using 20 ml sample in quartz vial indicated by  $\square$ .

of having a symmetrical system. If any difficulty arises in the electronics, it can be quickly pinpointed by the systematic substitution of symmetrical components. Incidentally, the low voltage power supply in each unit (amplifier, high voltage power supply, scaler and analyzer) is self-contained, so that one may take an amplifier out of the main cabinet and test it individually on a laboratory bench. If the need ever arises, it is possible to replace each component quickly.

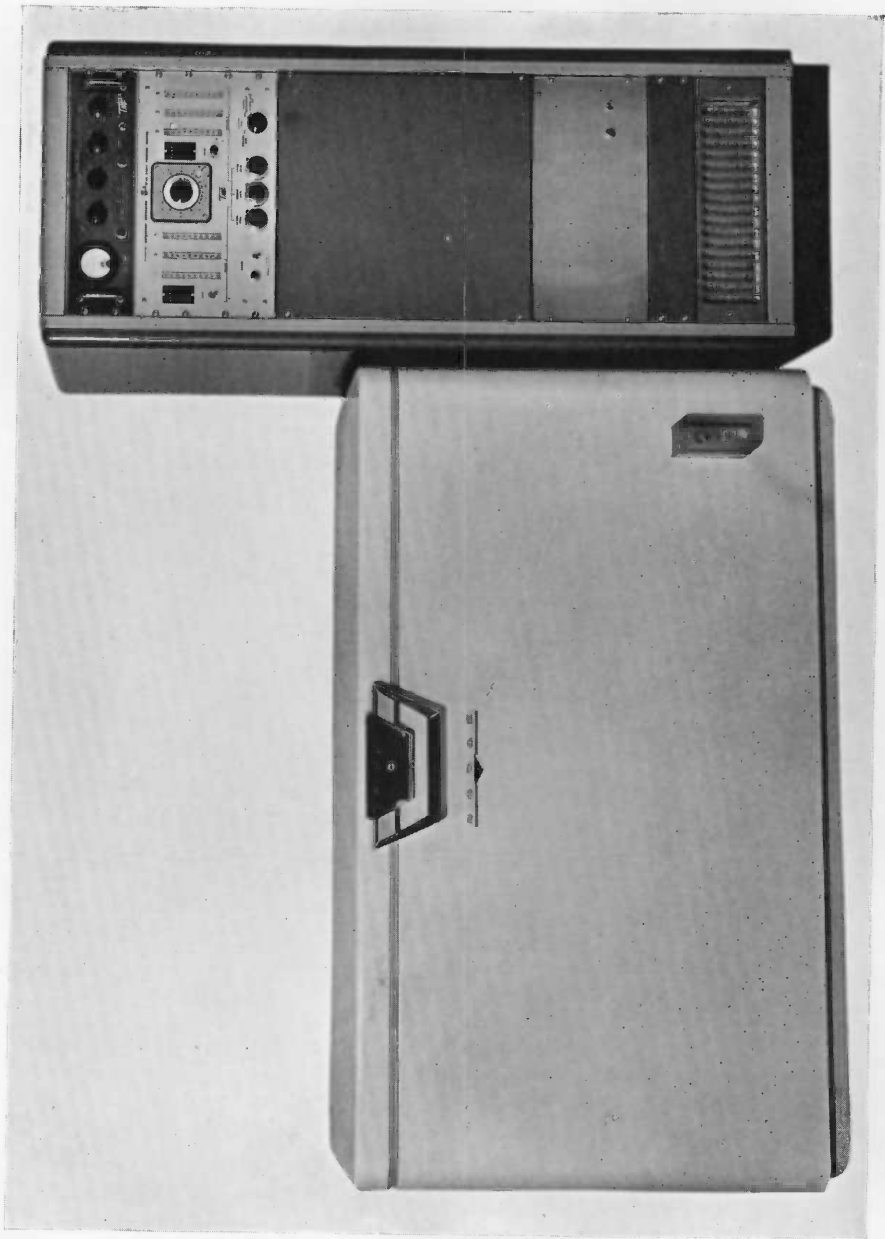


Fig. 4.

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The final performance of our liquid scintillation coincidence counter is shown in Figs. 2 and 3. The maximum carbon-14 efficiency is better than 90%, and the maximum tritium efficiency is 30%. The maximum efficiencies for carbon-14 and tritium using the optimum 1 to 6 differential discriminator settings are 55% and 20% respectively. Figure 3 shows an analysis of the background. The majority of the irreducible activity appears to be potassium-40 activity in the glass of the photomultiplier tubes. This activity scintillating in the glass envelopes has an energy peak closely corresponding to that of tritium, so that it makes up little of the background contribution when carbon-14 is counted. Potassium-40 contained in the sample vials is another normal source of background. When the counter is operating at 0°C, the accidental count due to the coincidence resolving time of the pulse analyzer is approximately 2 counts/min.