

A TRIPLE SAMPLE  
LIQUID SCINTILLATION COUNTER

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ABSTRACT

One of the major limitations of present liquid scintillation counters is their inability to count more than one sample at a time. Sample throughput would be greatly enhanced if two or more samples could be counted at the same time in one instrument. Functional parts of the liquid system could be shared, rather than duplicated, resulting in significant economies.

A liquid scintillation counter is described which measures three liquid scintillation samples simultaneously. Three phototube detectors are used to measure the three samples in a shared coincidence counter. Detection efficiency and background count rate are comparable to single sample counters. Isolation between the three sample chambers is better than 100 parts per million.

Four potential sources of error particular to analyzing events produced in a shared multiple detector system are examined: 1) pulse pile up, 2) dead time, 3) accidental coincidence, and 4) cross coupling between detectors.

INTRODUCTION

An important measure of liquid scintillation counting performance is the rapidity with which an assay can be performed. Most applications in the clinical and biological research laboratory require rapid throughput of relatively high activity samples. Improvements in detection efficiency and sample transport time would reduce counter time little. Except for tritium, most isotopes count at nearly 100% efficiency and transport time is typically less than

10 percent. In the environmental, health physics, and geological applications low activity samples are prevalent.  $E^2/B$  is an important figure of merit when the activity of the sample being assayed is equal to, or less than, the background rate (1). However,  $E^2/B$  is also a measure of minimum time to assay an unknown.

In either case an instrument which measures three samples simultaneously has counting efficiency and background rate essentially equal to that of all three samples added together. Whether measured by counting time, efficiency, or  $E^2/B$ , performance is improved by nearly a factor of three over the single sample counter. This paper describes a liquid scintillation counter which conserves time by counting three samples simultaneously.

## MATERIALS AND METHODS

### Detector

A new detector, using three 75mm phototubes to measure three samples, is shown top view in figure 1. The three samples are separated by a "Y" partition in the prism-shaped optical cell. Scintillations produced in chamber A are visible to multiplier phototube (MPT) 2 and MPT 3, but not MPT 1. Similarly, scintillations produced in chamber B are visible to MPT 1 and MPT 3, but not MPT 2. Three coincidence detectors, one for each sample, are used to separate the scintillations from each chamber. Similarly three summed amplifiers are used to analyze the pulse height spectrum from each chamber.

The detector head is constructed of 48mm steel plates. Each phototube is mounted in a steel housing (not shown) which slips into one of three ports on the side. Lead bricks, 5 cm thick, surround the detector (excluding phototubes).

The prism shaped cell is constructed of 1.5mm steel plates welded and machined for dimensional stability. Over all dimensions are 9.5cm on each side by 7.7cm high, sufficient to accommodate three 20 ml counting vials. Each sample chamber is lined with specular aluminum to minimize optical losses due to absorption. The "D" shaped opening from each chamber to the phototube has an area of  $12\text{cm}^2$  (78% of a 50mm tube).

Phototubes are EMI type D247B, 75mm diameter with 10

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stages of SbCs venetian blind dynodes. Each bialkali (K-Cs) photocathode has an effective area of  $33\text{cm}^2$ , twice that of the 50mm tube. Since the photocathode is shared by two counting chambers, less than half is available after partitioning. The cathode to dynode 1 inside surface of each phototube is coated with an anti-reflection coating to reduce the optical coupling between chambers. The smooth flat faceplates are made of 4mm Pyrex glass.

The interface between the "Y" shaped optical barrier and the phototube faceplate is designed to absorb light reflecting off the cathode from one chamber to another (figure 2). The black rubber seal is optically coupled to the faceplate. Because of the 4mm thickness,  $T$ , of the faceplate, the barrier seal is 7mm wide ( $D$ ) losing over  $3\text{cm}^2$  of cathode area.

### Detector Logic

Coincidence gating has been an accepted method of reducing background in a liquid scintillation counter for over 20 years (2). Thermionic emissions from the photocathodes of the phototube detector are virtually eliminated by imposing the requirement of coincidence. Although each phototube may produce several thousand thermionically generated output pulses per minute, the chance of a simultaneous occurrence in both is rare. On the other hand, each scintillation from a beta decay produces many photons within several nanoseconds, some of which are detected, almost simultaneously, by each phototube. The coincidence detector is arranged to have a time window,  $\tau$ , sufficiently wide (30 nanoseconds) to accept pulse pairs which are not exactly simultaneous.

Three coincidence detectors A, B and C, one for each chamber, are required to properly locate the origin of the scintillation in the three sample system (figure 5). Scintillations in chamber A excite MPT 2 and MPT 3. Each event will produce, almost simultaneously, pulses at both inputs to coincidence detector A and therefore produce an output from coincidence detector A. Coincidence detectors B & C will not produce an output because neither will be energized at the #1 input. All three coincidence detectors are identical. Each has a detection threshold of approximately ten microamperes and a resolving time,  $\tau$ , of about 30 nanoseconds.

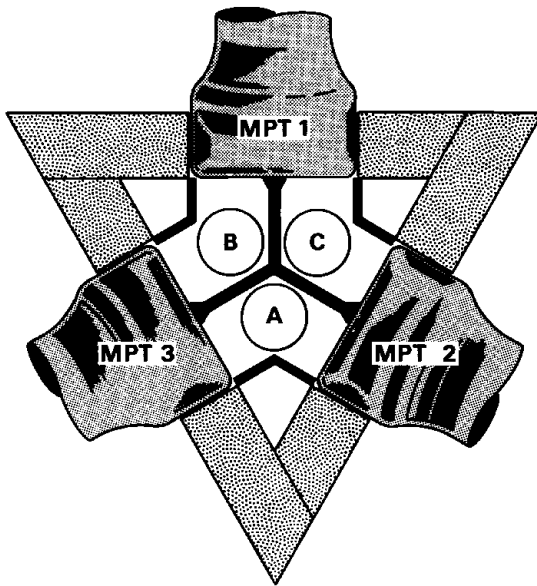


Figure 1 Detector partitioning.

MULTIPLIER PHOTOTUBE NO. 1

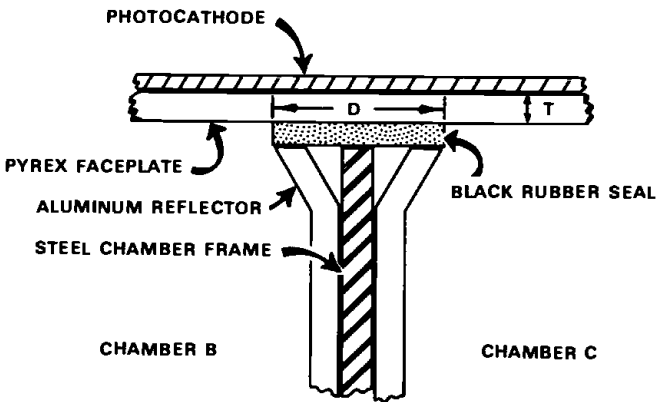


Figure 2 Cross-sectional view of optical barrier between counting chambers.

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A first differential pulse height discriminator (3) was employed to discriminate between light emissions from the phototubes (4) and scintillations produced by the sample. This circuit imposes a requirement of symmetry on the detector. Light generated in the sample tends to produce equal output from both phototubes and is accepted, whereas light generated asymmetrically, as within the phototube, is rejected because it exceeds the threshold of the Differential Discriminator. The differential threshold increases linearly with pulse height to accommodate any pulse height pair.

The Differential Discriminator was redesigned for the three sample system to operate upon three inputs. Rather than triplicate the Differential Discriminator function (one for each sample), it was modified to always respond to the largest and median pulse height from the three phototubes, independent of the phototube pair. Thus, the same differential discriminator criteria is automatically applied to the two phototubes with the largest pulse amplitude. Signals are also derived from this circuit to identify which phototube pair is the largest.

When two of the samples are high in activity the probability of chance simultaneous disintegrations from both is increased. If the scintillations from both samples are sufficient to excite all three phototubes simultaneously, the origin of the events is uncertain. Furthermore, a pulse would be added to all three sample channels. Fortunately, these occurrences are rare and represent a small fraction of the total count rate. They, therefore, can be rejected and neglected to be accounted for with little loss in system accuracy. The Triple Coincidence Detector shown in Figure 5 is used to inhibit all three outputs at "AND" gates A, B and C.

Optical isolation between chambers is imperfect. Intense scintillations from a sample in chamber A have increasingly good probability of exciting MPT 1 because of reflections from the inner surfaces of the phototubes. In an experiment to determine the amount of optical coupling, a  $^{36}\text{Cl}$  liquid scintillation reference source was positioned in chamber A, and the spectrum from chamber C was recorded (figure 3). Chamber B was filled with opaque material to prevent coupling via MPT 3, chamber C contained a blank sample, and the Triple Coincidence and Differential Discriminator were disabled.

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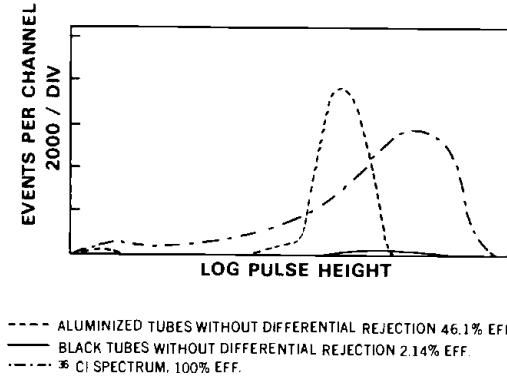


Figure 3  $^{36}\text{Cl}$  Cross coupling spectra without triple coincidence rejection.

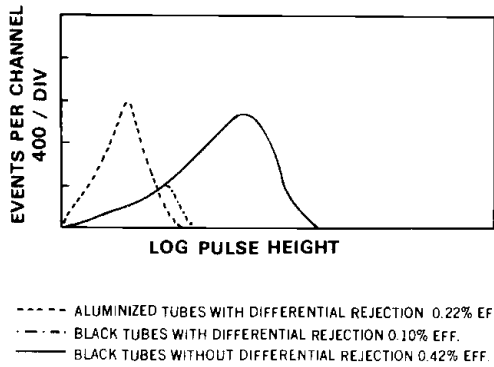


Figure 4  $^{14}\text{C}$  Cross coupling spectra without triple coincidence rejection.

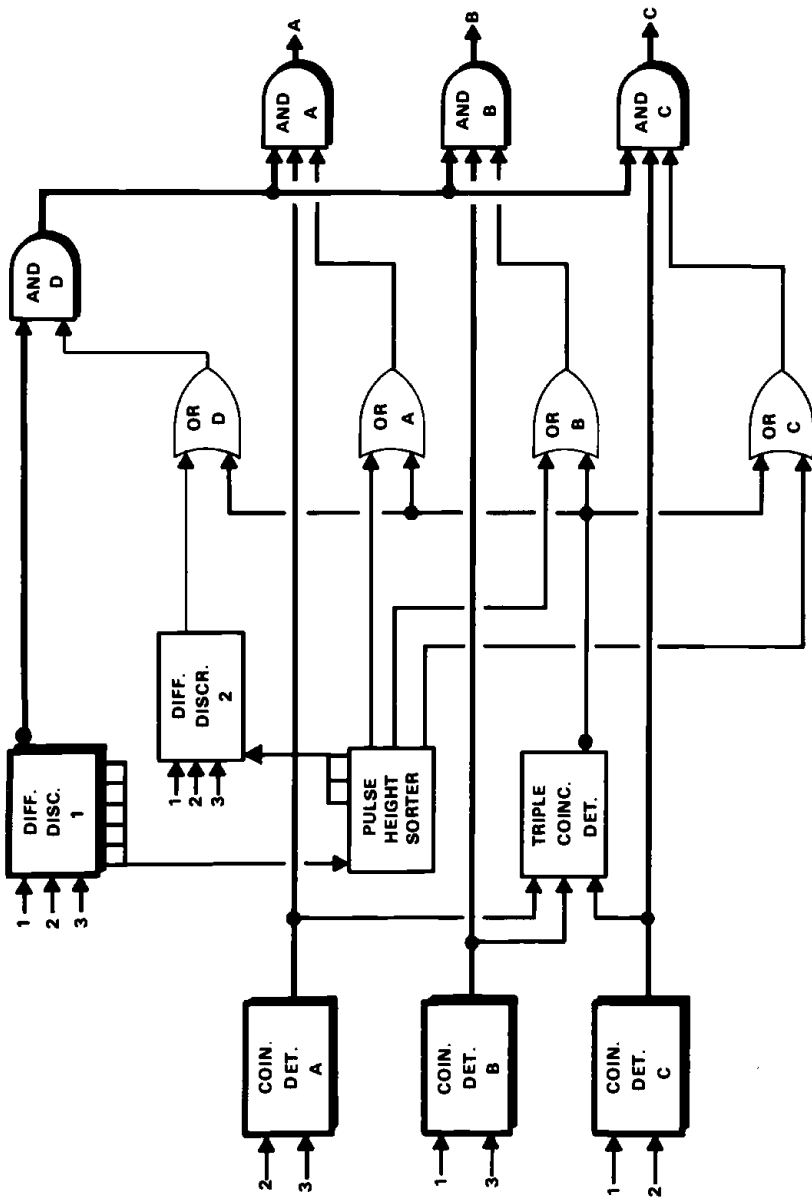


Figure 5 Triple sample detector logic diagram.

The spectrum from each chamber is the summed pulse height from the two adjacent phototubes. Since little light from the sample reaches MPT 1, the cross coupling spectra shown in figures 3 and 4 are essentially the spectra of events from MPT 2 which are detected by MPT 1. Although the antireflection coating significantly reduced the optical cross coupling, an additional method was required to reduce it to a negligible error.

In figure 4, the  $^{36}\text{Cl}$  source was replaced with a  $^{14}\text{C}$  source to examine the effectiveness of a Differential Discriminator to identify cross coupling. The original 2 input version of the Differential Discriminator was used. The sharp drop in the cross contribution spectra above 100 photons (about 1/3 of the log scale) shows that a Differential Discriminator can effectively identify virtually all cross contributions above 100 photons.

Because of the imperfect optical isolation, an intense scintillation can excite all three phototubes and be rejected by the Triple Coincidence Detector. A Pulse Height Sorter and a second Differential Discriminator are provided to override the triple anticoincidence, when legitimate scintillations excite all three phototubes. The Pulse Height Sorter is required because the coincidence logic is unable to determine the chamber of origin. Signals derived from the first Differential Discriminator are used to determine the chamber of origin. The most likely chamber of origin is opposite the phototube with the lowest pulse amplitude. The Pulse Height Sorter activates one of the three "OR" gates A, B or C. These are steering gates which determine which of the chambers is most likely to have produced the scintillation. The quantitative decision as to sufficient probability of being a legitimate event is determined by the second Differential Discriminator. It determines if there is sufficient difference between lesser and median pulse heights, to be reasonably sure the triple coincidence was caused by optical crosstalk, not by random coincidence of two events. The second Differential Discriminator is directed to examine the lesser and median amplitude pulses by the Pulse Height Sorter.

In summary, the detector logic consists of five elements to identify and accept or reject each scintillation from the detector. Spontaneous thermal electrons from the photocathode are rejected by the three coincidence detectors. Spontaneous luminous emissions from within the phototubes

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are rejected by Differential Discriminator 1. Occasionally random coincidences of events from different samples are rejected by the triple anti-coincidence circuit. Scintillations of undeterminable origin which excite all three photocathodes are also rejected by the triple anti-coincidence circuit. Scintillations with sufficient intensity to excite all three phototubes are located by the Pulse Height Sorter. Differential Discriminator 2 determines if there is sufficient probability that the location determined by the Pulse Height Sorter is sufficiently correct to accept the event. Very few triple coincidences are indeterminate.

## RESULTS

Tritium detection efficiency averaged 51.3% in the three chambers, about 13% (absolute) lower than in a single sample detector. The Carbon 14 and chlorine 36 efficiencies measured 95.4 and 100% respectively. The difference is attributable to chamber optics. The optical cell efficiency can be improved by the addition of a reflector around the top of the sample vial and by minimizing the area of black seal which is exposed. A reflecting top surface with a round hole for the sample, as is used in conventional detectors, would add about 10% more reflector surface to the chamber. Elimination of the black rubber seal exposed around the circumference of each window in the chamber would add another 15% to the reflecting surface.

Background count rate measured about 20 cpm greater than in a conventional detector. Total integral background averaged 62.9 cpm; 23.8 cpm in the  $^3\text{H}$  energy range (unquenched) and 17.6 cpm between the  $^3\text{H}$  and  $^{14}\text{C}$  end-points. About 8 cpm can be removed in the  $^3\text{H}$  energy range by replacing the Pyrex face plates with low background material, such as quartz (5).

Chamber isolation was determined by placing an active sample in one chamber and measuring the cross-contribution detection efficiency in each of the other two chambers. Blank samples were placed in the other two chambers and background was subtracted from the gross count rate to obtain the net cross-contribution. Tritium, Carbon 14 and chlorine 36 cross-contribution efficiency measured 0.005%, 0.0015% and 0.0012% respectively. Cross-contribution is

less at higher energies because both the triple anticoincidence and differential discrimination functions are more effective at higher energies.

## DISCUSSION

### Pile Up and Dead Time

Potential errors in pulse height analysis, due to pulse pile up and dead time losses, are slightly more than in a single sample counter. The essential difference is that the activity of all three samples contribute to the error. Conventional pulse amplifier and linear gating techniques can virtually eliminate pulse pile up as a source of error. Similarly, conventional live timing can eliminate potential errors caused by analyzer dead time. Because of the additional dead time, it takes a little more time to count three samples simultaneously than one at a time. The difference becomes significant only at high count rates.

Assume each sample has  $10^6$  cpm and the average dead time per event is 2 microseconds. The triple sample counter is busy analyzing events 10% of the time whereas the single sample counter is busy only 3% of the time. Therefore, the triple sample detector will take 7% longer to count three  $10^6$  cpm samples than to count one.

Accidental coincidences can either increase or decrease the count rate in a triple sample detector. A decrease occurs when events are discarded by the triple coincidence detector. Let us distinguish between double and triple accidental coincidences.

### Double Accidentals

The probability of an accidental double coincidence event for any phototube pair is the same as in a single sample detector:

$$A_2 = 2\tau S_1 S_2$$

where  $\tau$  is the coincidence resolving time for each input to the coincidence detector (30 nanoseconds).  $S_1$  and  $S_2$  are the single phototube count rates. The singles rates include chemiluminescence and phosphorescence excitation in addition to thermionic emission.

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Present day alkali phototubes have thermal emissions typically from 1 to 20 thousand counts per minute and produce negligible accidental coincidence. However, chemiluminescence and phosphorescence samples may easily produce hundreds of thousands of counts per minute at each phototube. Decreasing the resolving time,  $\tau$ , only linearly diminishes the accidental count rate; whereas the accidental count rate increases with the square of the luminescent rate.

In a triple sample detector a second type of double accidental coincidences can occur. High monophotonic event rates can cause not only an erroneous increase in count rate but can indicate the wrong sample. Assume chamber A is empty and there are two samples in chambers B and C with intense monophotonic event rates. Each random coincidence between a pulse at MPT 3 and MPT 2 caused by samples B and C respectively, will appear as a coincidence from the empty chamber A. These events will not be rejected by the Triple Coincidence Detector because MPT 1 is unexcited during these chance coincidences. It is best to avoid high chemiluminescence or phosphorescent samples or to wait until the luminescent energy has decayed to an emission rate which produces negligible accidental coincidences.

Low energy scintillations can produce the same kind of counting error when the intensity of the scintillations are low enough to excite one but not both phototubes. The maximum probability of an accidental coincidence in the wrong detector is less than 0.01% even when both samples have an activity of  $10^6$  disintegrations per minute at the worst intensity, 3-5 photons. Less intense scintillations are less likely to produce any detectable output. More intense scintillations are more likely to be rejected as triple coincidences.

### Triple Accidentals

Triple coincidences may occur due to optical crosstalk between chambers or from the accidental coincidence of events in two different samples. Triple coincidences caused by optical crosstalk are identified by the Pulse Height Sorter and Differential Discriminator 2 and correctly counted. Accidental triple coincidences cannot be properly analyzed and are rejected. Although two legitimate events are discarded the fractional loss is small. Even if

all three samples have count rates of  $10^6$  cpm the loss amounts to only 0.2% assuming a coincidence resolving time of 30 ns.

The probability of an accidental triple coincidence is:

$$A_3 = 2\tau(R_1R_2 + R_2R_3 + R_1R_3)$$

where  $A_3$  = accidental triple coincidence rate

$R_1, R_2, R_3$  = count rates in each chamber

However, each triple coincidence rejected is a loss of two events for all three samples. Therefore the loss per sample is 2/3 the accidental coincidences (assuming equal count rates).

#### Gamma Emitters

Samples containing gamma emitting radionuclides can produce cross coupling between chambers. Scintillations from Compton electrons may be produced in one sample by a gamma emitted from another. Cross coupling from a low energy gamma emitter like  $^{125}\text{I}$  (35keV) can be virtually eliminated with 1.5mm of lead shielding. However, shielding becomes unpractical for gammas above 100keV. Because of the required close proximity of the samples little shielding can be added to protect against cross coupling from high energy gammas.

A triple chamber detector is a practical method of improving sample throughput. Improvements in detector optics are required to achieve performance equal to that of current single sample detectors. Data obtained from any sample is virtually independent of the activity or beta energy of adjacent samples. Gamma emitters below 100keV may also be assayed with negligible cross-contribution.

#### ACKNOWLEDGMENT

The author wishes to thank Mr. H. Engberg and Mr. J. M. Dudley for their assistance particularly in analyzing the sources of cross-contribution

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