Chapter 2

Recent Developments in Photomultipliers for Liquid Scintillation Counting

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INTRODUCTION

Considerable new interest is being expressed in the field of liquid scintillation counting. This interest exists due to increased efforts in the clinical, research and environmental monitoring areas. The latter is of principal concern because of the increase in tritium-producing nuclear power facilities. Attendant with this interest there has been an introduction of new photomultipliers for use in liquid scintillation counters.

This paper discusses a new photomultiplier for liquid scintillation counting, the 4501V4. It is contrasted with the generic type 4501V3 in terms of E^2/B in the tritium and carbon windows, crosstalk, background and accidental count rates. A discussion of the effect of tube materials upon photomultiplier performance is given. Future trends in photomultipliers are also discussed.

PHOTOMULTIPLIER DISCUSSION

The 4501 photomultiplier types employ the Matheson-type photocathode-to-first dynode ('front end') geometry[1] coupled to a 12-stage BeO, focussed electron-multiplier structure. The anode output structure consists of a grid-type anode and three-element, parallel plane transmission line. The main features are shown diagrammatically in Fig. 1.

The 2-in diameter spherical-section front end affords two advantages over alternative planar faceplate designs:

1. The photoelectron trajectories are nearly isochronous, and the electric field strength near the photocathode is higher, affording faster time response, and thus a narrower time coincidence window for improved noise rejection.
2. The spherical section permits a reduced mass of faceplate material which in turn provides a lower background from the photomultiplier.

A bialkali (K—Cs—Sb) photocathode is used which has a quantum efficiency of approximately 31% at 385 nm. The 12 BeO dynodes provide typical gains of 1.4×10^7 at 2 kV applied voltage. Anode pulse risetime for delta-function excitation of the photocathode is approximately 2.5 ns, affording time response that exceeds the requirements of most present-day liquid scintillation counting systems.

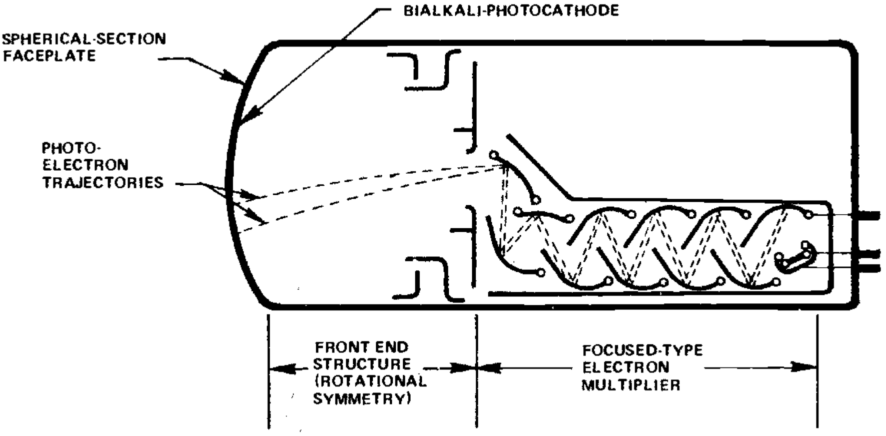

Fig. 1. Diagrammatic representation of 4501 photomultiplier

The focussed electron-multiplier structure has the following advantages over venetian blind-type multiplier structures which are also used in photomultiplier detectors:

1. The time response of the focussed multiplier is typically an order of magnitude faster than that of venetian blind multipliers.
2. The focussed multiplier has a higher total collection efficiency than the venetian blind multiplier. Coates reported a collection efficiency of ~95% for a focussed structure multiplier (8850, which is generically similar to a 4501V4) against 66% for a venetian blind multiplier (9558QB).[2]

MATERIALS CONSIDERATIONS

The envelope and internal tube parts materials chosen for photomultipliers used in liquid scintillation counting applications are very critical. One obvious requirement is that all tube materials shall exhibit negligible radioactivity. Of prime concern in this regard is the glass used in the vacuum envelope housing the photomultiplier.

The glasses most commonly used in photomultiplier construction contain some ^{40}K and thorium daughter-products. These radioactive contaminants are found in the raw materials comprising the glass, such as sand, and are also introduced into the glass when it is melted in thoria refractories.[3] The former problem can be overcome by judicious monitoring of the basic constituents prior to melting, and the latter problem can be solved by proper choice of melting vessel, platinum being an ideal choice.

Low background counting experiments performed on glasses from manufacturers throughout the world indicate a considerable spread in radioactive content. Some data on activity of glasses used in photomultipliers are presented in Table 1.

Less well understood, but of equal importance, is the fluorescent activity of the faceplate glass used in the photomultiplier. All glasses scintillate to some extent when excited by X-rays or γ-rays, and such scintillations lead to undesired background counts in the case of liquid scintillation counters. A pronounced scintillation-like effect due to cosmic ray bombardment is commonly seen in photomultipliers.[5,6] Unpublished measurements at this facility indicate that commonly used photomultiplier window materials vary by several orders of magnitude in their scintillation efficiencies. The problem of scintillating windows can be approached from at least two avenues:

1. Employ special window materials with very few luminescent centres ($<1:10^9$), or
2. Add dopants to common window materials to quench the luminescence (or shift it to a sufficiently long wavelength such that it is of no consequence).

Both approaches have been tried in photomultipliers with good success.

Table 1. Radioactive content of some glasses commonly used in photomultiplier construction (activity is stated in counts/min-kg for 2π counting geometry and a 70 keV to 2.6 MeV energy window).

Glass type	Manufacturers	Activity (counts/min-kg)
7740	Corning Glass Works, Corning, N.Y.	83
KG-33	Owens-Illinois, Vineland, N.J.	53
7070	Corning Glass Works	290
7050	Corning Glass Works	171
EN-1	Owens-Illinois	567
7052	Corning Glass Works	465
7720	Corning Glass Works	208
0080	Corning Glass Works	69

Data from A.W. Consylman and D.D. Crawshaw.

It is clear that considerable improvements can be made to photomultipliers by incorporating the best possible choice of materials in their construction. This approach has been followed in the 4501V3 and 4501V4 photomultipliers, as will be described in the next section.

TUBE EXPERIMENTS

The experiments were performed in a commercially available liquid scintillation spectrometer.[7] Data are reported on a large (>200) tube sample, ensuring statistically valid average values. In all experiments the tubes were 'paired' in the sense that tubes with comparable gains were used together. This requirement stemmed from the limited gain—adjust capability of the instrument (a characteristic of nearly all commercially available spectrometers), and was therefore an instrument-imposed constraint.

Unless otherwise stated, all measurements were made in the tritium energy window and with a tritium counting efficiency of 60%. The carbon counting efficiencies for carbon-above-tritium measurements averaged approximately 70%. The figure-of-merit E^2/B (efficiency, in percent, squared, divided by background, in counts per minute) is stated, in keeping with common usage.

The crosstalk measurements were made with the spectrometer elevator down and with an empty counting chamber. The background measurements were made with a Packard background standard. The accidental count rate was measured with the elevator shaft blocking the tubes.

To prove the magnitude of the cosmic ray/Cerenkov effect, certain measurements were repeated with the entire spectrometer rotated 90° so that the photomultiplier axis was vertical instead of horizontal.

Results and interpretation of experiments

Table 2. Results of experiments comparing 4501V3 with 4501V4.

Parameter	4501V3	4501V4	Remarks
Tritium E^2/B (60% eff.)	160	210	
Background in tritium window (c.p.m.)	22.5	17.1	Measured with Packard standard
Crosstalk in tritium window (c.p.m.)	12	7	
Accidental rate (c.p.m.)	0.8	0.8	Measured with elevator shaft blocking tubes, calculated value ~0.07 c.p.m.
Carbon-above-tritium E^2/B (70% eff.)	563	546	A difference was noted between the pulse height distribution of the background between the V3 and V4 tubes in the carbon energy window

The results of the experiments are tabulated in Table 2. In brief, the E^2/B has been increased from 160 to 210 in the 4501V4 as contrasted to the 4501V3. It should be noted that this increase was achieved through a reduction in background (22.5 c.p.m. to 17.1 c.p.m.) and crosstalk (12 c.p.m. to 7 c.p.m.). From these data one may conclude that the material-related improvement in the 4501V4 was due largely to a reduction in scintillation efficiency of the window, as opposed to a reduction in radioactivity of the envelope, as the background standard is an efficient scintillator.

To assess the magnitude of cosmic ray interactions, the crosstalk and accidental rates were measured with the spectrometer in the horizontal (normal operating) position and in the vertical position. This experiment was performed on only one pair of tubes, owing to the mechanical difficulties of rotating the spectrometer. The data are summarised in Table 3.

The crosstalk in the tritium window increased from 5.7 to 8.6 c.p.m. in going from horizontal to vertical, a ratio of 1.5. The accidental rate increased from 0.34 to 0.95 c.p.m. horizontal to vertical, a ratio of 2.8. The higher ratio for the accidental rate is ascribed to the increased probability of an energetic particle interacting with both tubes while the system is vertical.

The accidental rate may be calculated from the relationship $A = 2 N_1 N_2 \lambda$, where N_1 and N_2 are the dark count rates of the two individual photomultipliers and λ is the resolving time of the coincidence system. Typical values in our experiments were $N_1 = N_2 \sim 10 \times 10^3$ c.p.m. and $\lambda = 20$ ns, giving $A = 0.07$ c.p.m. The order-of-magnitude increase in measured accidental rate over calculated rate was attributed to cosmic ray events, as demonstrated by the greatly increased accidental rate for vertical operation of the system.

Table 3. Effect of cosmic rays upon spectrometer orientation — 4501V4

	Spectrometer horizontal (normal position)	Spectrometer vertical	Remarks
Crosstalk in tritium window (c.p.m.)	5.7	8.6	Measured on one pair of tubes
Accidental rate (c.p.m.)	0.34	0.95	Made on one pair of tubes

FUTURE IMPROVEMENTS

Further improvements can be made to photomultipliers to reduce their background and fluorescent properties, and research is being carried out in this area. Additional refinements can also be made to systems to improve performance with existing photomultipliers.[8] A future possibility would be a single tube liquid scintillation counter utilising pulse shape and pulse height discrimination to eliminate the need for a two-tube coincidence system. Such a system with a suitable photomultiplier could distinguish between tube dark pulses (single-electron in origin), tube Cerenkov pulses (fast pulses of multiple-electron origin) and scintillator signal pulses (relatively slower pulses of multiple-electron origin) on the basis of pulse height and pulse shape (time) information. Photomultipliers such as the developmental type C31024 may have the basic time response and electron resolution capabilities to render such a system feasible in today's state-of-the-art.

SUMMARY

Materials used in photomultiplier construction have been investigated for radioactive and fluorescent properties. The results of this investigation have led to the introduction of the 4501V4 photomultiplier which offers a tritium E^2/B of 210 as opposed to 160 for the present 4501V3 type. Further improvements in these photomultipliers are being researched, and possibilities of a single tube liquid scintillation spectrometer utilising pulse shape and pulse height discrimination are being studied.

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DISCUSSION

P.E. Stanley: Do you have any data on the amount of light reflected at the quartz/photocathode interface and does this vary with the wavelength of the incident light and the thickness of the photocathode?

D.E. Persyk: We do not have the data to answer that question.

P.E. Stanley: What order of variation in quantum efficiency (at a given wavelength, say 380 and 425 nm) do you observe? What I'm really after is the variation in response across the photocathode. Would it be of the order ± 20% of the observed quantum efficiency?

D.E. Persyk: Yes, it's of that order.

T.A. Wilkins: With reference to Table 2 (Comparison of Performance of P.M. Tubes 4501V3 and 4501V4), were the results indicated subjected to analysis of variance and statistical tests? These yield (1) further information of validity of results, (2) reproducibility of manufacture of tubes.

D.E. Persyk: Yes, the results were subjected to routine analysis of variance and statistical relevance. Photocathode processing of the tubes is done by a process-control computer which also performs an on-line test of each tube's important performance parameters. The computer processing affords excellent reproducibility, which is important as the tubes are matched in liquid scintillation counting applications.

T.A. Wilkins: Some of the differences between measurements on different tubes were very small, whilst errors on these could be quite large. No errors were presented nor were any confidence levels presented with respect to significance of differences.

D.E. Persyk: The values are cited with an uncertainty in the least significant digit to a confidence level of 68%. For example, in Table 2 background in the tritium window for the 4501V4 is shown as 17.1 c.p.m. This was obtained with a 1000 minute count so that the measurement results could be expressed more precisely as 17.1 c.p.m. ± 0.1 c.p.m. The authors stress that the key point regarding the statistical relevance of these measurements is not the small uncertainty associated with each measurement, but rather the large (greater than 200 tube) sample size that was employed.

R.P. Parker: What is the speed of response of your photomultipliers? Whilst a single photomultiplier system is excellent in terms of the physics of the system, can chemiluminescent events be discriminated against using pulse-shape discrimination?

D.E. Persyk: Rise time is 1 to 2 nanoseconds, 800 picoseconds at best. Chemiluminescence will be an important problem and we have not investigated this. Chemiluminescence (and phosphorescence) is also a problem in a conventional two-tube coincidence system. The lower level discriminator is set to ignore single-electron events, which are associated with chemiluminescence, owing to the single photon nature of the process. However, at high count rates pulse pile-up gives rise to false counts. This condition can be recognised by unbalancing the system: A delay line having delay greater than the coincidence window time width is inserted in one leg of the coincidence circuit — then, if the count rate does not change appreciably, the counts are attributed to pulse pile-up from chemiluminescence. In a single-tube system the lower level discriminator is likewise set at the one-electron level, and therefore discriminates against chemiluminescent signals up to some maximum count rate. The presence of false counts due to chemiluminscence could be recognised by a count rate circuit which monitors the single-electron rate and issues a warning when an unacceptably high signal rate is detected.