A SINGLE PHOTOMULTIPLIER LIQUID SCINTILLATION COUNTING APPARATUS FOR ^{14}C LOW-LEVEL MEASUREMENT

Fu Pei-yun and Zhang Ting-kui
(Institute of Biophysics, Academic Sinica)
Beijing, China

A single low noise photomultiplier (PMT) liquid scintillation counting apparatus used for ^{14}C low level measurement has been developed. Samples in quartz vials are counted on the end window of a PMT. In addition to 10 cm of lead shielding, two kinds of anticoincidence shielding were tested; a G-M counting tube and a liquid scintillation solution. With the liquid scintillation solution as an anticoincidence shield, the ^{14}C efficiency for 5 ml samples was more than 70% and the background was 1.42 cpm. If some silicone oil was introduced between the sample vial and the end-window of the PMT the collection efficiency of light was improved. Under this condition, the background was 0.62 cpm with the efficiency approximately 70%, resulting in a figure of merit (E^2/B) of 7,900 for a 5 ml sample.

This single photomultipler apparatus can operate stably without cooling at room temperature (24°C).

INTRODUCTION

The earliest liquid scintillation counter used one photomultiplier tube to measure the fluorescence produced in a liquid scintillator. The background of these instruments was very high because of the high noise from the PMT. Cooling techniques were used to decrease the noise. In 1957, W. Bernstein et al.[1] reported a single tube apparatus used for ^{14}C counting. Although cooling was adopted, the background was as high as 80 cpm. Since the coincidence technique has been used in liquid scintillation counting this situation has been considerably improved. Because of the development of bialkali photochathodes, quartz windows and the improved manufacturing technology of PMTs the background from PMTS has been greatly decreased, and it is now possible to design a single tube apparatus for ^{14}C low background measurement[2].

On the basis of researching and manufacturing low-level liquid scintillation counters,[3,4] we designed and built a single photomultiplier (EMI 9635QB) liquid scintillation counting apparatus without cooling. Two kinds of anticoincidence shield, G-M counting tube and liquid scintillation solution were tested. The results show that the anticoincidence effect of liquid scintillator is better than that of G-M tube. If some silicone oil is introduced between the sample vial and the end-window of the PMT, the efficiency (E) for measuring a 5 ml sample (in a quartz vial) is over 70% with a background (B) below 1 cpm and the figure of merit (E^2/B) approaches 7,000. Without silicone oil, the figure of merit approaches 5,000.

APPARATUS

Besides the anticoincidence shield and PMT, this apparatus is similar to the older types. We used an anticoincidence shield to reduce the background. Two kinds of shield counter, G-M tube and liquid scintillation solution, were tested. Fig. 1 and Fig. 2 show the detector part of the apparatus.

Fig. 1 shows the structure of the apparatus shielded by a hollow anode G-M tube (Model J-309, Shanghai Electron Tube Factory). The main detector is a low background EMI 9635QB PMT selected from several tubes. The G-M tube covers the sample vial. Both detectors are then surrounded by 10 cm of lead.

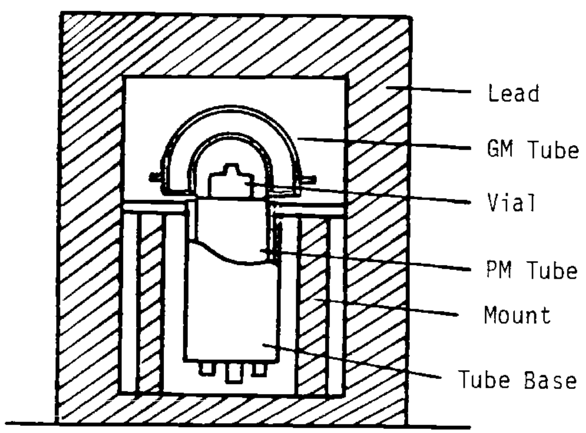

Figure 1. Detector Assembly with GM Anticoincidence Shield.

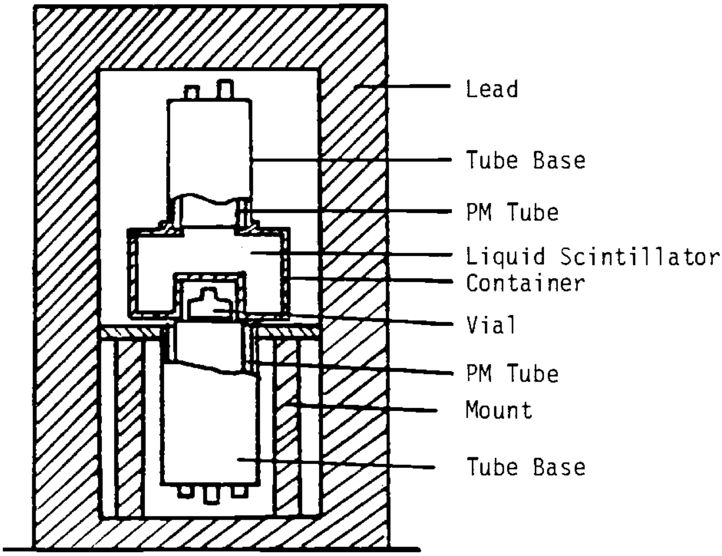

Figure 2. Detector Assembly With LS Anticoincidence Shield.

Fig. 2 shows the structure of this apparatus shielded by a liquid scintillation counter. The main detector is the same as shown in Fig. 1. A stainless steel container is used for the liquid scintillator (5 g PPO + 0.1 g POPOP/l Toluene). The thickness of the scintillator above the sample vial is about 8 cm, and that around the side of the vial is about 5 cm.

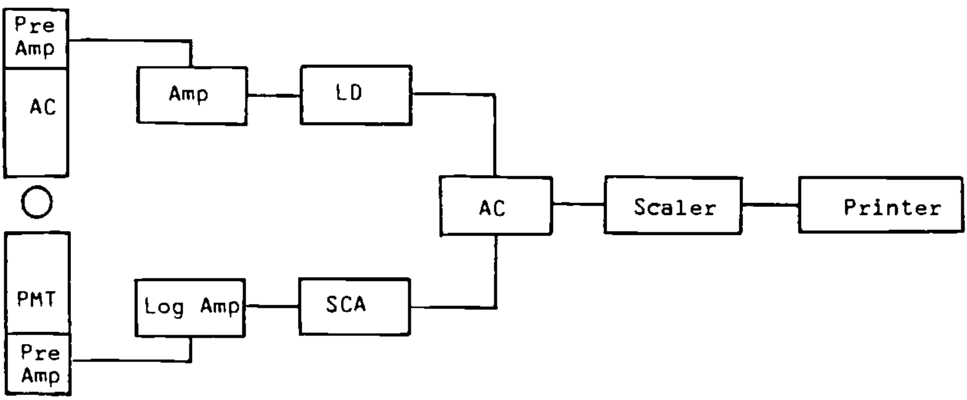

Figure 3. Block Diagram.

RESULTS

The main sources of background in the liquid scintillation counter can be divided into three parts[5]: cosmic ray and surrounding activity, the noise of the PMT and the radioactivity in the material around the sample chamber. In this single PMT apparatus, some experiments concerning light collection and shielding were undertaken. The effect of lead shielding on the background is very significant. When the thickness of lead is increased from 5 cm to 10 cm, the background is approximately reduced by 44% or 65% depending on the kind of anticoincidence shield. The experimental results are shown in Table 1. The composition of scintillator is 6g/l Butyl-PBD in Benzene.

In a coincidence liquid scintillation counter "cross-talk" is an important source of the background. Both electronics and optics can be used to reduce "cross-talk"[6,7]. But in the single tube apparatus, these methods are ineffective. A sodium iodide crystal, organic scintillator and GM counting tube have been used in liquid scintillation counting as an anticoincidence counter, but most of these were used in coincidence type LS counters. We chose two kinds of them: hollow anode GM counting tube and liquid scintillation solution. The hollow anode tube is very cheap. It has a solid angle of more 2 pi, so the anticoincidence effect is quite good. When a liquid scintillation solution is used, another PMT is needed. So two PMTs are used. In this case, if the figure of merit were not

Table 1. The effect of lead thickness on the background. (Quartz sample vial, 5 ml).

lead (cm) anticoincidence	5 cm	10 cm	E (%)	B (cpm)
G-M	5.15 ± 0.07	2.87 ± 0.05	>70	2.28
LS	4.04 ± 0.05	1.42 ± 0.03	>70	2.62

Table 2. The efficiency and background with and without anticoincidence shield. (Quartz vial, 5 ml).

anti-coincidence method	E (%)	without anti-coin.		with anti-coin.	
		B (cpm)	E^2/B	B (cpm)	E^2/B
G-M	74	9.45 ± 0.15	580	2.87 ± 0.05	1900
LS	74	8.00 ± 0.10	680	1.42 ± 0.03	3800

obviously better than that of the coincidence type counter, the additional complication of our approach would not be warranted. But the result in Table 2 is inspiring. Using a liquid scintillator as an anticoincidence counter is more expensive than using G-M tube, but its performance is better. The effects of the two kinds of anti-coincidence shield on the background are shown in Table 2.

The background of the single tube results mainly from discharge, field emission and natural radioactivity. The lower the applied high voltage, the less discharge and field emission are observed. Therefore, for ^{14}C counting lower voltage on the PMT is preferred. It is appropriate to choose the high voltage at which the integrated efficiency of ^{14}C is about 80%. Under this condition, most of the PMT noise diminishes, because the ^{14}C signals correspond to more than two photoelectrons in the PMT. If the efficiency for collecting the fluorescence from the sample is improved, more photoelectrons can be produced. Then the effect of the single tube background in the lower energy region on ^{14}C spectrum can be decreased. The noise can be eliminated by an appropriate setting of the lower threshold of the ^{14}C counting window.

In general, it is an effective method of improving light collection to include some silicone oil between the sample vial and the end-window of the PMT. When the silicone oil is used, the energy

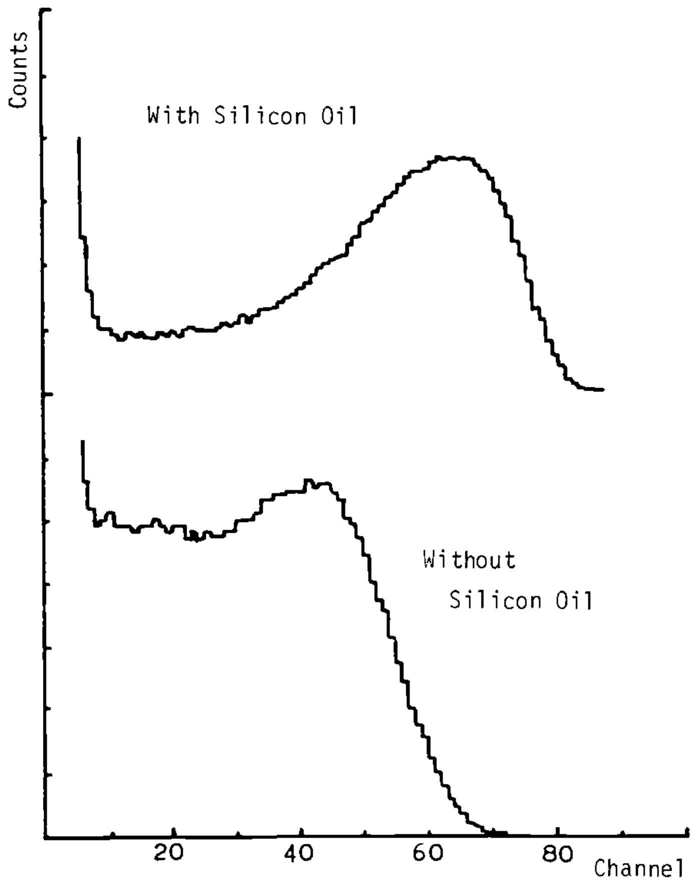

Figure 4. The Effect of Silicon Oil on ^{14}C Spectra using an Aluminum Cylindrical Reflector.

spectrum of ^{14}C moves to a higher amplitude region of the single tube background spectrum (Fig. 4). The low threshold of a counting window can thus be raised so that the contribution of the background from the PMT itself will be decreased.

In Table 3, data are compared under two conditions, with and without silicone oil.

Table 3. The effect of silicone oil on performance (26°C) (quartz vial, 5 ml).

Sil. Oil	E (%)	without AC B (cpm)	with AC B (cpm)	E^2/B
without	73.7	8.00 ± 0.10	1.42 ± 0.03	3800
with	70	7.36 ± 0.09	0.62 ± 0.02	7900

In our single PMT apparatus, the light collecting efficiency is affected by the shape of sample chamber. The truncated cone with teflon lining is better than an aluminum cylinder (see Fig. 5). Fig. 6 shows the ^{14}C spectrum for these two chambers with the same operating voltage. Table 4 shows the efficiency and the background for these two systems.

Table 4. The effect of the shape of sample chambers on performance. (5 ml quartz vial, without silicone oil).

Shape	E (%)	B (cpm)	E^2/B
cylinder	73.7	1.42 ± 0.03	~ 3800
truncated cone	71.5	0.99 ± 0.02	~ 5100

When silicone oil is used as a light coupler, even if a higher efficiency of ^{14}C is chosen, the background is not very high. Table 5 shows the experimental results with 5 ml quartz vial under different operating conditions.

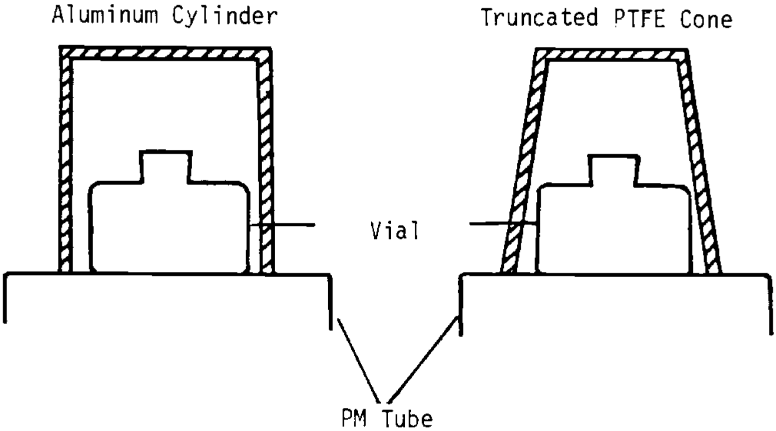

Figure 5. Two Shapes of Sample Chamber.

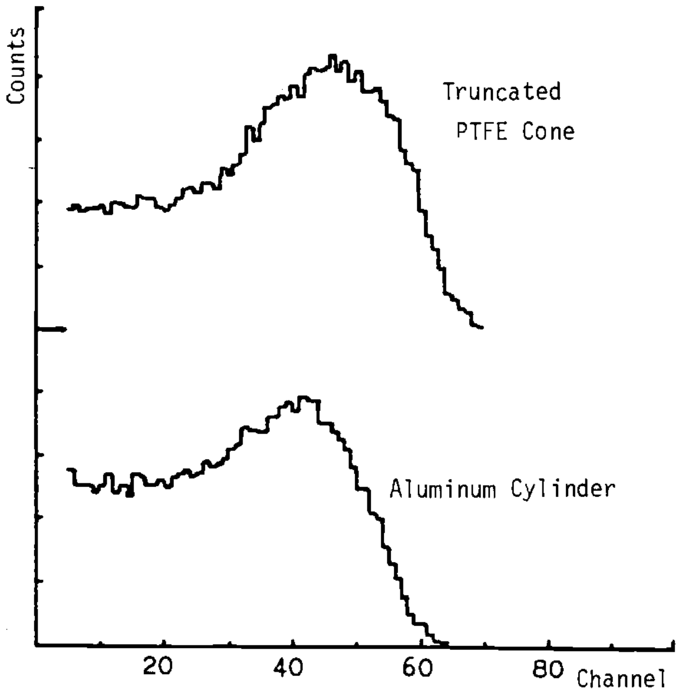

Figure 6. The Effect of Sample Chambers on ^{14}C Spectra (Without Silicon Oil.)

Table 5. Performance under different operation conditions. (H.V. 900 V).

Window	1.7 to 2.4	1.4 to 3.0	1.2 to 3.0	0.9 to 4.0
E (%)	70	80	85	89
B (cpm)	0.66	1.00	1.14	1.42
E^2/B	7500	6400	6300	5500

We have tested several PMTs (EMI 9635QB) with different noise performance. From Table 6, the effect of single-photoelectron noise on background for ^{14}C can be hardly seen. When the background for a ^{14}C window is 0.62 ± 0.02 cpm, the background due to the PMT in the ^{14}C window, i.e., without a sample, is 0.21 ± 0.01 cpm. Thus, it can be seen, the background in the ^{14}C window is mainly caused from the light produced in the PMT and the sample by cosmic ray and other external radiation.

Table 6. The effect of PMT noise level on ^{14}C background (5ml, quartz vial).

PMT Serial No.	Noise (cpm)	without sil. oil E (%)	without sil. oil B (cpm)	with sil. oil E (%)	with sil. oil B (cpm)
27020	1265 (1450 V)	73.7	1.42 ± 0.03	72.3	0.70 ± 0.02
26693	5715 (1600 V)	72.3	2.29 ± 0.05	74.4	1.05 ± 0.03
22033	37598 (1500 V)	73.0	4.84 ± 0.07	73.0	1.55 ± 0.03
26704	5816 (1800 V)	73.2	1.29 ± 0.03	73.9	0.87 ± 0.03

In general, the performance of a PMT is affected by ambient temperature. But in our apparatus, the variation of the efficiency and background of ^{14}C (Table 7) caused by variations in ambient temperatures is not apparent. This proves that the performance of this single PMT apparatus is adequate to satisfy the demands of low-level ^{14}C counting. Furthermore, it proves that single PMTs can be used for low-level ^{14}C counting at the room temperature without cooling.

The contribution of sample vials to background can be divided to two main parts: from the Cerenkov light excited by cosmic rays and other radioactivity in the vial wall, and also from radioactive impurities in the vial material. Table 8 shows that due to the contribution of ^{40}K, the background of even low potassium glass vials is higher than quartz vials. However, low potassium glass vials could be used because they are cheaper and their background is not that much higher.

Table 7. Long term stability of ^{14}C efficiency and background (without silicone oil, 5 ml sample).

Date	Temperature (°C)	E (%)	B (cpm)	E^2/B
Jan. 14-17	14.5	72.6	1.03 ± 0.03	5100
Feb. 4-6	14.5-12	72.7	0.96 ± 0.02	5500
Feb. 21-22	10-11	73.1	1.01 ± 0.03	5300
Mar. 8-10	16-15	72.5	1.03 ± 0.02	5100
Apr. 11-12	16-18	71.6	1.02 ± 0.03	5020

Table 8. Low Potassium Glass Vial

No.	4	6	8	10	13
B (cpm)	2.09 ± 0.04	2.59 ± 0.05	1.86 ± 0.04	1.94 ± 0.05	2.16 ± 0.04

Quartz Vial

No.	1	3	4	6	10
B (cpm)	0.88 ± 0.03	0.87 ± 0.03	0.93 ± 0.03	0.92 ± 0.03	1.03 ± 0.02

DISCUSSION

The single photomultiplier tube liquid scintillation counter with anticoincidence shield that we have built is suitable for ^{14}C low-level counting. There are several advantages: good performance, reduced cost, ease of construction and freedom from cooling. It has all the potential for application in many regions. Due to the noise of PMTs, however, this apparatus is not suitable for tritium low-level counting without cooling.

From ^{14}C spectra and background spectra we observed that the main source of background in a ^{14}C window is the light produced by external radiation. With an anticoincidence shield, we can eliminate most of the background in the ^{14}C window caused by this source.

It is simpler and cheaper to use the hollow anode counting tube as an anti-coincidence counter, but its life is limited. Although the liquid scintillator is more expensive, it is cheaper than NaI(Tl) or a plastic scintillator, and has the advantage of long life. In particular, the shielding effect of a liquid scintillator is better than that of G-M tube.

REFERENCES

1. Bernstein W., Bjerknes C. and Steele R. "Single Phototube Liquid Scintillation Counting of ^{14}C" in "Liquid Scintillation Counting" Bell C.G. and Hayes F.N. (Eds.), Pergamon Press, New York, pp. 74-77, 1958.
2. Rajamae R and Punning JM "Some Possibilities for the development of ^{14}C measurements by liquid Scintillation Counting" Radiocarbon, 22, 2, pp. 435-441, 1980.
3. Jiang Han-ying "Model DYS Low Level Liquid Scintillation Counter" Liquid Scintillation Spectrometer Group, Institute of Biophysics, Biochemistry and Biophysics, 5, p. 68 (in Chinese) 1982.
4. Jiang Han-ying "Model DYS Low Level Liquid Scintillation Counter" Liquid Scintillation Spectrometer Group, Institute of Biophysics, Nuclear Electronics and Detection Technology, 2, 5, p. 8 (in Chinese) 1982.
5. Allessio M, Allegri L, Bella F and Improta S. "Study of the Background Characteristics by Means of a High Efficiency Liquid Scintillation Counter" Nuclear Instruments and Methods 137, pp. 537-543, 1976.
6. Laney BH "Electronic Rejection of Optical Crosstalk in a Twin Phototube Scintillation Counter" in "Organic Scintillators and Liquid Scintillation Counting" Horrocks DL and Peng CT (Eds.), Academic Press, New York, pp. 991-1003, 1971.
7. Noakes, JE "Considerations for Achieving Low Level Radioactivity Measurements with Liquid Scintillation Counters" in "Liquid Scintillation Counting" Crook MA and Johnson P (Eds.), vol. 4, Heyden, London, pp. 189-206, 1977.