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PERFORMANCE OF THE PACKARD 2000 CA/LL AND 2250 CA/XL LIQUID SCINTILLATION COUNTERS FOR ^{14}C DATING

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ABSTRACT. Results are presented on the comparative characteristics of the Packard 2000 CA/LL liquid scintillation counter with and without the low-level option on line. An initial performance assessment using ^{14}C labeled benzene with butyl-PBD as the scintillant revealed that although the background count rate decreased by approximately a factor of 3 using the low-level option, a substantial decrease in efficiency was also observed. However, subsequent investigations have indicated that, by careful manipulation of both scintillant composition and concentration, this loss in efficiency can largely be overcome with little or no concurrent increase in background count rate. The introduction of an active vial sample holder and a new light-guide system proved to be significant advances on the standard 2000 CA/LL.

INTRODUCTION

Liquid scintillation counters are generally used in bio-medical research because of the widespread applications of β-emitting radioisotopes. The counters are produced relatively inexpensively and in large numbers. Their features are not ideally suited to low-level counting applications such as ^{14}C dating, groundwater (^{3}H) dating and general environmental monitoring for contamination from nuclear industry.

Genuine low-level scintillation counters, a more recent innovation, generally employ anticoincidence (active) shields as well as perhaps more massive passive shielding to reduce background count rates, enabling improved signal-background ratios which are acceptable for low-level applications.

The Packard 2000 CA/LL liquid scintillation counter brings a completely novel approach to scintillation counting, providing low-level counting with relatively inexpensive systems that require no active or enhanced passive shielding nor special laboratory conditions. The 2000 CA/LL approach is to discriminate unquenchable background events (which represent ca 2/3 of the total background) from organic scintillation events (quenchable) (which represent the remaining 1/3 background events) together with genuine β disintegrations from the sample by means of pulse shape/duration analysis. A major criticism of this counter is that, although the background count rate is significantly reduced, so also is efficiency. Polach *et al* (1988) concluded that the optimum ^{14}C efficiency obtainable from the Packard 2000 CA/LL for a 3ml geometry was 54.3% (1.01 cpm background) in a standard 7ml slimline glass vial, while an efficiency of 76.4% (0.21 cpm background) was achievable from the Pharmacia-Wallac Quantulus using a 3ml teflon/copper vial.

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Since the Packard concept depends totally on discriminating short-duration true β events (1-2n sec) from longer duration non-quenchable background events (up to 5μ sec) (van Cauter, 1986), then it would seem logical that a proportion of the true β events are being rejected because their pulse widths are greater than the cut-off threshold defined within the software. Other than reprogramming this discrimination software, the most obvious possibility for regaining the observed losses in efficiency would be to sharpen the pulse widths from true β events.

EXPERIMENTAL

Two ^{14}C benzene standards were used during this study. The first, of 178.5 ± 2.0 dpm.g^{-1} activity, was prepared at SURRC. The second was that used by Polach *et al* (1988) and corresponds to 25.7 ± 0.2 dpm.g^{-1}; this already contains 15mg.g^{-1} butyl-PBD (2-(4-Biphenylyl)-5-(4-tert-Butyl-phenyl)-1,3,4-Oxadiazole). The first stage in this experimental program was to establish optimum concentrations of the primary scintillants butyl-PBD and PPO (2,5 Diphenyloxazole). The criteria for establishing optimum concentrations were constancy of efficiency as measured on a 0-156 keV window and constancy of quenching obtained from the t-SIE value. t-SIE is the spectral index of the transformed Compton spectrum of the external standard. The t-SIE range is 0-1000, a value of 1000 indicating no quenching. The study by Polach *et al* (1988) used butyl-PBD at both 6.8mg.g^{-1} as recommended by Packard for the 2000 CA/LL and 17.1mg.g^{-1}, presumably an optimum for the Quantulus.

Second, these optima were assessed in the presence of the secondary scintillants, bis-MSB (p-bis-(σ-Methylstyryl)-Benzene), POPOP (1,4-bis-[2-(5-Phenyloxazolyl)]-Benzene), and dimethyl POPOP (Me_2 POPOP) (1,4-bis-[2-(4-Methyl-5-Phenyloxazolyl)]-Benzene; bis-MSB was also assessed alone.

Third, the second of the two ^{14}C standards (25.7 dpm.g^{-1} containing butyl-PBD) was reassessed, both in its original form and with the most effective secondary scintillant, to determine the optimum E^2/B and $No/\sqrt{B}$ values. The lower discriminator was kept at the ca 100% ^3H cut-off point and no higher. Subsequently, this sample was then assessed in conjuction with an active plastic scintillator vial holder and, finally, in the Packard 2250 CA/XL counter with a new plastic scintillator light-guide system. Finally, a selection of scintillant combinations were assessed in parallel in the 2000 CA/LL and 2250 CA/XL counters for efficiency, quenching and background on a 0-156 keV window. Stages 1, 2 and 4 were carried out using 4.5g benzene in a standard 20ml screw-cap glass vial (Packard). Stage 3 employed the corresponding 7ml slimline vial. Counting times for the higher activity standard were typically 100 mins, yielding an overall efficiency error of < 0.4%. Background counting times were typically 500-1000 min, while for the low activity standard, > 300 min.

RESULTS AND DISCUSSION

Table 1A shows that, in the low-level counting mode, open-window efficiency decreases from 84.1% to 74.8% as the concentration of butyl-PBD increases from 2-14mg.g^{-1}. In parallel, however, quenching, as reflected by the t-SIE value, decreases with increasing scintillant concentration (2-12mg.g.$^{-1}$). In the conventional counting mode, the trend in t-SIE value is identical but efficiency is constant (ca 93%) from 4–20mg.g^{-1}. Although 6.8mg.g^{-1} butyl-PBD is recommended by Packard, the results presented here indicate that this is far from optimum. The loss in efficiency is not perhaps as great as with higher scintillant concentrations; however, it is within a region of both quenching and efficiency instability as reflected by the major influence of scintillant concentration on both the t-SIE and efficiency values. The results presented here indicate that a minimum of 12mg.g^{-1} is required for stable counting conditions; however, efficiency is reduced by ca 18-20% compared to the conventional counting mode. Table 1B likewise indicates spectral stability for PPO at ca 5.6mg.g^{-1}, while the loss in counting efficiency is much less than for butyl-PBD (ca 10%).

The addition of the secondary scintillants, POPOP and Me_2 POPOP (Table 2) had no influence on counting efficiency in conjuction with PPO in the low-level counting mode, while in conjunction with butyl-PBD, they improved efficiency only marginally (ca 4%) (Table 3). The addition of bis-MSB increased the efficiency obtainable from both PPO and butyl PBD (Tables 2, 3), enabling low-level counting efficiencies of 88-90% compared

TABLE 1

Influence of the concentration of the primary scintillants butyl-PBD and PPO on ^{14}C spectral stability and counting efficiency with and without the low-level option on line

Scintillant (mg.g^{-1} of C_6H_6	Eff (%) L L* on	t-SIE** L L on	Eff (%) L L off	t-SIE L L off	Difference in % eff
A. Butyl-PBD					
2	84.1	557	91.8	557	7.7
4	80.7	683	93.5	681	12.8
6	78.9	730	92.8	732	13.9
8	76.6	749	93.5	751	16.9
10	75.7	757	93.2	755	17.5
12	75.2	761	93.6	762	18.4
14	74.8	761	93.1	760	18.3
16	73.9	760	93.4	759	19.5
18	73.5	758	93.0	759	19.5
20	73.7	755	93.8	755	20.1
B. PPO					
2.2	85.4	582	92.5	579	7.1
3.3	84.8	620	91.7	614	6.9
4.4	84.0	635	92.8	629	8.8
5.6	83.6	639	93.5	635	9.9
7.8	82.8	638	92.6	634	9.8
10.0	82.3	634	92.6	629	10.3

*Low-level option = pulse shape discriminator
**Spectral index of external standard

TABLE 2

Effect of incorporating varying concentrations of secondary scintillants (POPOP, Me_2 POPOP, and bis-MSB) into a fixed concentration of PPO (5.6mg.g^{-1} of C_6H_6)

Secondary scintillant (mg.g^{-1} of C_6H_6	Eff (%) L L on	t-SIE L L on	Eff (%) L L off	t-SIE L L off	Difference in % eff
A. POPOP					
0	83.6	639	93.5	635	9.9
0.1	83.9	652	93.5	651	9.6
0.2	82.9	661	93.8	659	10.0
0.7	81.9	696	93.8	696	11.9
1.1	82.0	694	93.4	688	11.4
B. Me_2 POPOP					
0.1	83.2	615	92.8	613	9.6
0.2	82.3	632	93.4	630	11.1
0.7	82.6	653	93.6	652	11.0
1.1	82.4	671	92.7	663	10.3
C. Bis-MSB					
0.2	85.4	661	93.0	659	7.6
0.7	87.5	682	93.1	682	5.6
1.3	88.0	695	93.8	696	5.8
2.7	89.6	702	93.7	699	4.1
5.3	89.1	672	93.7	678	4.6

TABLE 3

Effect of incorporating varying concentrations of secondary scintillants (POPOP, Me_2 POPOP, and bis-MSB) into a fixed concentration of butyl-PBD (12mg.g^{-1} of C_6H_6)

Secondary scintillant (mg.g^{-1} of C_6H_6	Eff (%) L L on	t-SIE L L on	Eff (%) L L off	t-SIE L L off	Difference in % eff
A. POPOP					
0	74.6	760	94.0	762	19.4
0.1	76.6	767	94.5	768	17.9
0.2	76.3	770	93.7	770	17.4
0.7	78.7	767	93.2	766	14.5
1.1	78.8	759	94.0	759	15.2
B. Me_2 POPOP					
0	75.1	755	93.5	755	18.4
0.2	75.6	732	93.6	734	18.0
0.7	77.2	726	93.5	726	16.3
1.3	78.1	726	94.0	725	15.9
2.7	78.8	715	93.5	717	14.7
C. Bis-MSB					
0	74.7	757	93.3	754	18.9
0.2	81.1	752	93.5	755	12.4
0.7	83.9	752	93.9	750	10.0
1.3	86.6	743	93.6	740	7.0
2.0	87.4	734	93.5	732	6.1
2.7	87.4	722	93.2	718	5.8
3.3	88.0	708	93.3	707	5.3
4.0	88.5	696	93.1	695	4.6
4.7	87.2	680	92.7	681	5.5
5.3	88.0	679	93.6	683	4.6

TABLE 4

Influence of the concentration of the secondary scintillant bis-MSB alone on ^{14}C spectral stability and counting efficiency with and without the low-level option on line

bis-MSB ($mg.g^{-1}$ of C_6H_6)	Eff (%) L L on	t-SIE L L on	Eff (%) L L off	t-SIE L L off	Difference in % eff
0.7	87.2	383	89.1	382	1.9
1.3	88.9	533	91.6	532	2.7
2.7	89.0	602	91.8	602	2.8
5.3	88.5	637	93.0	637	4.5

TABLE 5

Progressive optimization of the Packard low-level counting system using an improved scintillant, a plastic scintillant vial holder, and a new plastic scintillant light guide system (3ml geometry in slimline glass vial)

Counting conditions	Background	Eff(%)	E^2/B	$No/\sqrt{B}$
1) Butyl-PDB ($17.1mg.g^{-1}$)in Packard 2000 CA/LL	1.03	52.5	2680	16.8
2) Butyl-PBD ($17.1mg.g^{-1}$) with bis-MSB ($1.1mg.g^{-1}$) in Packard 2000 CA/LL	1.25	63.8	3250	18.5
3) As in 2) with plastic vial holder in Packard 2000 CA/LL	0.92	66.1	4750	22.4
4) Butyl-PBD ($17.1mg.g^{-1}$) with bis-MSB ($1.1mg.g^{-1}$) in Packard 2250 CA/XL	0.91	64.6	4580	21.4
5) As in 4) with plastic vial holder in Packard 2250 CA/XL	0.83	65.5	5170	22.7

with 93-94% under conventional conditions. Similarly, bis-MSB alone (1.3-5.3$mg.g^{-1}$) (Table 4) also enabled counting efficiencies of ca 89%.

When the low activity standard was reassessed as per Polach *et al* (1988) (Table 5), the results obtained for optimum performance were largely comparable in terms of both efficiency and background. The addition of bis-MSB enabled an increase in optimum efficiency from 52.5 to 63.8% with, however, some increase in background (1.03 to 1.25 cpm). E^2/B and $No/\sqrt{B}$ values were nevertheless substantially increased. A further enhancement was then achieved using the active vial sample holder. By direct comparison, the Packard 2250 CA/XL produced a better overall response than the 2000 CA/LL, although slightly poorer than the latter counter used in conjunction with the active vial sample holder. The 2250 CA/XL in combination with the active vial sample holder produced the best overall response.

Table 6 compares the counting characteristics of a selection of scintillant combinations in both the 2000 CA/LL and 2250 CA/XL. The addi-

TABLE 6

Comparison of efficiency, quenching, and background between the Packard 2000 CA/LL and 2250 CA/XL for several scintillant combinations

Scintillant	2000 CA/LL			2250 CA/XL		
	Eff (%)	t-SIE	Bkgd	Eff (%)	t-SIE	Bkgd
12mg.g^{-1} butyl-PBD	75.2	761	8.26	6.1	760	2.59
5.6mg.g^{-1} PPO with 0.7mg.g^{-1} POPOP	81.9	696	7.69	80.5	799	4.48
5.6mg.g^{-1} PPO with 2.7mg.g^{-1} bis-MSB	89.6	702	8.74	89.6	807	5.20
12mg.g^{-1} butyl-PBD with 4mg.g^{-1} bis-MSB	88.5	696	8.68	88.1	808	5.34
5.3mg.g^{-1} bis-MSB	88.5	637	8.99	88.8	779	5.00

for PBD and PPO/POPOP are significantly lower than for the other scintillants, although, as already shown, total counting efficiency also decreases. With the exception of PBD alone, there is no difference in efficiency between the two counters. The efficiency from PBD is exceptionally low (6.1%) and is probably due to absorption of that wave length of light by the plastic scintillator. t-SIE values for PBD are unchanged, while for all other scintillants, values are increased by > 100 units in the 2250 CA/XL. From the results presented here, backgrounds for the three scintillant combinations giving optimum counting efficiency are not significantly different but show ca 40% decrease in the 2250 CA/XL.

CONCLUSIONS

Our results show that the Packard 2000 CA/LL is sensitive to both scinntillant composition and concentration. However, by careful manipulation of both parameters, it is possible to obtain counting efficiencies in the low-level counting mode almost comparable to those from conventional counting, within 4-5%, without substantially increasing the background count rate. One fact difficult to grasp is that, in some circumstances, efficiency increases as quenching increases. This obviously reflects the complexity of the pulse-shape analyses and must involve a complex balance of sufficient scintillant, self-absorption and energy transfer to scintillants capable of sharpening pulse widths. The results of this study merely touch on the work involved in optimizing this counter. Obviously, much work is required, perhaps at a much more fundamental level, eg, examining pulse shapes in relation to different scintillant compositions and then reprogramming the pulse discrimination software in light of optimized concentrations.

The introduction of the active vial sample holder and the new light-guide system are substantial advances on the original 2000/LL. Both counters represent a significant advance in ^{14}C dating by liquid scintillation and although, at present, their performance cannot match that of the Quan-

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tulus, they do have the advantage of being more routinely usable and not requiring special vials or greater passive shielding. The results for the 2250 CA/XL were obtained within one week, the counter having been optimized within 24 hours of receipt.

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