ELECTRONIC OPTIMIZATION OF SCINTILLATION COUNTERS FOR DETECTION OF LOW-LEVEL ^{3}H AND ^{14}C

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ABSTRACT

The performance of scintillation spectrometers in the hands of twenty-one leading low-level counting specialists is evaluated. The figure of merit varies from poor to excellent; this can be attributed to selection of counting vials and scintillation cocktails and to changes in operator selected counting parameters, rather than equipment type. This paper deals with electronic factors which lead to user or counter manufacturer accessible modifications. For test purposes, an LKB-Wallac Rack Beta counter was chosen. Its performance, before and after modification, is compared to four other counters. Factors evaluated are: characteristics of multiplier phototubes (MPTs) and based on these we postulate selection criteria for improved low-level counting of ^{14}C and ^{3}H; effects of high voltage reduction on ^{3}H and ^{14}C counting efficiency and background; effect of cross-talk and its partial suppression by masking of MPTs and by making changes in the acceptance ratio of the singles pulse-height comparator prior to pulse summation (an LKB-Wallac feature); reduction of coincidence resolution time; and increase in coincidence bias (as opposed to increase in low-level discriminator setting). The above investigations are illustrated graphically. Results attest to their merit. Sources of noise interfering with low-level counting have also been investigated. It was found that non-random events are generated by: very fast high voltage transient peaks transmitted along the supply line; radiofrequency (RF) pickup; vial and/or sample changer movement and handling generated static electricity discharge. The modified LKB-Wallac counter incorporates all the electronic modifications described, and is switch selectable for optimal ^{3}H or ^{14}C, and combined ^{3}H + ^{14}C low-level counting. The built-in noise

suppression features include a static suppressor (a high voltage low frequency discharge unit) and RF signal pickup whose amplified signal is channelled to inhibit the coincidence pulse of the PMs. The HV transient suppressor remains optional. However, a multi-channel pulse height analyzer interface is incorporated for user convenience.

PERFORMANCE OF LS COUNTERS

The most common way of comparing performance of liquid scintillation (LS) counters is given by the term Figure of Merit (FM) expressed as Ef^2/B, where Ef = % efficiency and B = background in counts per minute (cpm). Polach[1] recommended the use of a Relative Factor of Merit (F) for low-level counting where $F = No/\sqrt{B}$ where No = observed cpm of standard (^{14}C or ^{3}H) and B = background cpm. Its advantages are that F is related to volume of sample, which affects both No and B, whilst FM is sample volume insensitive. The usage of FM is inappropriate for low-level counting especially if the sample volume becomes small and/or is not specified. However, Ef^2/B still remains a useful term for other applications and we shall use it when relating to manufacturer specifications or when comparing relative merit of counters using the same vial and sample volumes.

The growing world-wide interest in Quaternary and Environmental research resulted in a significant increase in laboratories specializing in low-level research. The majority of these use LS spectrometers because simple yet quantitative sample benzene synthesis procedures are well documented in the literature and standard LS spectrometers, available 'off the shelf', perform reasonably well. We have attempted to find out how well by inviting some 20 low-level counting specialists to supply us with results which we present in Table 1. The bias towards ^{14}C is based on the specific interest we have at ANU. It is not representative of a world-wide trend although, it would be fair to say, that ^{14}C laboratories outnumber ^{3}H labs. Both require good low-level counting performance. However, only ^{14}C performance is compared in Table 1. All researchers who supplied us with results use benzene as their sample solvent and only 4 out of the 21 cited use the scintillator directly dissolved in the sample benzene. Others pre-dissolve it mostly in toluene. A number of scintillators are used, the most common is PPO. The most common

Table I. Relative performance of LS spectrometers in 21 laboratories specializing in low-level counting.

RESEARCHER	EQUIPMENT	MODI-FIED	COUNTING MATERIAL	VIAL SIZE (ml)	SCINTILLATOR & CONCENTRATION	COUNTING SOLUTION	COUNTING VOLUME (ml)	BKG cpm	.95 OX cpm	No/√B	EXTERNAL STANDARD	TRITIUM COUNTING
SCHRODER GEOL. SURVEY, DENVER	NUCLEAR CHICAGO M1	YES	Low K^{40}	8	1.0% PPO + 0.0025% dmPOPOP	1ml Tol + Scint + 3ml Sample Benz	4	11.1	25.7	7.7	NO	NO
REEBURGH UNI. OF ALASKA	PICKER 220	YES	Low K^{40}	10	0.5% PPO + 0.02% dmPOPOP	1ml Tol + Scint + 5ml Sample Benz	6	4.0	29.8	14.9	NO	NO
POLACH AUSTRALIAN NAT. UNI.	BECKMAN 200 (ROUTINE)	YES	Low K^{40}	5.5	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 4ml Sample Benz	5	5.5	30.8	13.1	NO	NO
POLACH AUSTRALIAN NAT. UNI.	BECKMAN 220 (EXPERIMENTAL)	YES	TEFLON	5.5	0.5% b-PBD(2)	5ml Sample Benz	5	2.0	40.8	28.8	NO	NO
				10.5		10ml Sample Benz	10	3.6	81.6	43.0	NO	NO
HARKNESS SCOTTISH UNI R.R.C.	PACKARD 3000 " 2425 INTERTECH SL30	?	Low K^{40}	20(1)	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 4ml Sample Benz OR	5	3.3	26.8	14.7	YES	NO
						2ml Tol + Scint + 8ml Sample Benz	10	4.8	50.0	22.8	YES	NO
BURLEIGH BRITISH MUSEUM	PACKARD 3315 " 3255	NO	Low K^{40}	20	0.4% PPO	10ml Tol + Scint + 5ml Sample Benz	15	7.5	35.2	12.9	YES	NO
MELHUISH INST. NUCLEAR SC.N.Z.	OWN DESIGN	N.A.	Low K^{40}	5.5	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 4ml Sample Benz	5	3.7	24.5	12.7	NO	NO
BUDDEMEIER UNI. of HAWAII	BECKMAN-100	YES	TEFLON	8	0.8% b-PBD	1.2ml Benz + Scint + 6.8ml Sample Benz	8	5.7	52.1	21.8	NO	YES
COLEMAN ILL. STATE GEOL. SUR.	PACKARD 3375	YES	Low K^{40}	10	1.0% b-PBD	2ml Tol + Scint + 8ml Sample Benz	10	5.2	64.8	28.4	NO	NO
TAMERS NOVA UNI.	PACKARD 2425	NO	Low K^{40}	5	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 3ml Sample Benz	4	7.0	22.3	8.4		
SCHARPENSEEL UNI. HAMBURG	PACKARD 3075 BERTHOLD 500	NO NO	QUARTZ	20(1)	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 4ml Sample Benz	5	7.0	35.9	13.6	YES	NO
GILLESPIE SYDNEY UNI.	PACKARD 2211	YES	Low K^{40}	5.5	0.4% PPO + 0.01% dmPOPOP	1ml Tol + Scint + 4ml Sample Benz	5	3.6	28.0	14.7	NO	NO

CALF	OWN DESIGN	N.A.	TEFLON	8	0.6% PPO(2)	5ml Sample Benz	5	1.6	28.7	22.7	NO	YES
AUSTRALIAN A.E.C.												
HAAS	INTERTECH-20	YES	QUARTZ	4	0.9% b-PBD	3.0ml Sample Benz	3	2.6	22.2	13.8	NO	NO
S.M.U. DALLAS			(SQUARE)	2.0		1.5ml Sample Benz	1.5	2.3	11.1	7.3	NO	NO
				0.8		0.6ml Sample Benz	0.6	2.1	4.6	3.2	NO	NO
				0.4		0.3ml Sample Benz	0.3	1.9	2.3	1.7	NO	NO
THOMAS, Jr.	PICKER-200	?	Low K^{40}	7	0.4% PPO +	1ml Tol + Scint	5	5.5	26.5	11.3	YES	NO
STATE UNI. N.Y.					0.01% dmPOPOP	+ 4ml Sample Benz						
DELLA	OWN DESIGN	N.A.	Low K^{40}		NE216 ? %	6ml Benz + Scint	10	6.9	38.5	14.7	NO	NO
UNI. of ROME						+ 4ml Sample Benz						
PUNNING	VACUTRONIC-GDR	NO	Al/Low K^{40}	13	0.4% PPO +	20% m-Xylene + Scint	12	4.8	60.0	27.4	YES	NO
ACAD. SCI. ESTONIA	? -USSR	NO	Al/Q(3)	11	0.01% dmPOPOP	80% Sample Benz	10	2.2	43.9	29.6	YES	NO
				22			21.5	3.2	108.3	60.5	YES	NO
OTLET	PACKARD 2425		Low K^{40}	22	NE231A	?	?	8.3	?	?	YES	YES
HARWELL	PACKARD 5375											
STIPP	BECKMAN-100	?	Low K^{40}	4.5	0.4% PPO +	1ml Tol + Scint	4	8.5	24.3	8.3	YES	YES
MIAMI	BECKMAN-200				0.01% dmPOPOP	+ 3ml Sample Benz		4.7	23.4	10.8		
	PACKARD-2003							5.3	23.7	10.3		
DRESSER	OWN DESIGN	N.A.	QUARTZ	13	62% NE216	7.5ml Tol + Scint	12.5(4)	3.4	36.1	19.6	NO	NO
UNI. COL. CARDIFF			FLAT F.			+ 5ml Sample Benz						
KENDALL	SEARLE	NO	Low K^{40}	2.5	0.8% b-PBD	2ml Sample Benz	2	2.5	19.3	12.2	YES	NO
UNI. of HONG KONG				5.0		5ml Sample Benz	5	3.9	37.5	19.0		
				10.0		10ml Sample Benz	10	7.8	73.9	26.5		

(1) = Vials are masked with aluminium foil or black paint above counting vol to reduce background

(2) = Scintillator weighed directly into sample C_6H_6

(3) = Aluminium vial with low K^{40} or quartz window

(4) = Nitrogen-flushed

counting volume is about 5 mL. The variation in $No/\sqrt{B}$ cannot be related directly to the type of equipment used except in the case of PUNNING (5th from bottom of Table 1) whose counter incorporates an active anticoincidence guard (gas counter) to reduce environmental radiation[2]. This lifts its performance well above average, Fig. 1. Almost all major manufacturers are represented. Many of the counters were home-modified (presumably to improve performance); some were home-built. We rate performance as poor if F = <10; as average if 11-20; above average if 21-50; and superior if >50 (Fig 1). We conclude that variations of performance can be attritubed to selection of counting vials, scintillation cocktails and to changes in operator selected counting parameters rather than equipment type. We note the superiority of the counter fitted with an anticoincidence device[2]. We also wish to stress that the survey is not comprehensive as there are some very notable omissions. The survey was meant to sample a cross section of common usage of LS counters for low-level counting. As such, it is representative and quite useful.

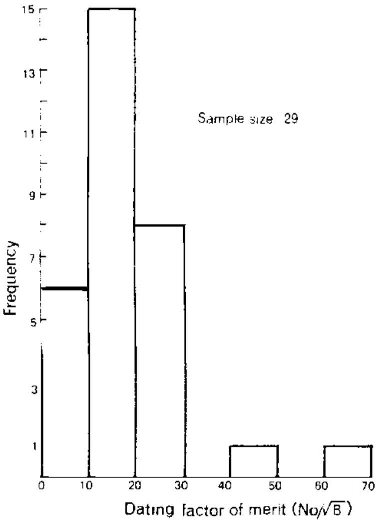

Figure 1. Distribution of counting merit, based on Radiocarbon Dating Factor ($No/\sqrt{B}$), achieved by 21 specialists.

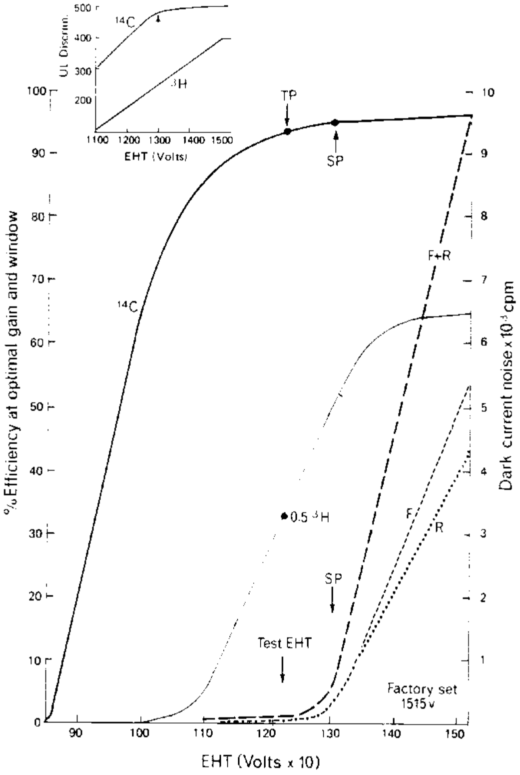

Figure 2. Efficiency of ^{3}H and ^{14}C detection and dark current noise as a function of EHT applied to the MPTs. SP is saturation point above which increase in ^{14}C efficiency is minimal, shift of relative pulse height end-point is nil (cf. inset) and dark current noise increases sharply. TP is recommended ^{14}C test point for highest No/√B. It corresponds ~ to 0.5 ^{3}H (32% Ef).

This paper will now deal with counting parameters which can be influenced electronically.

Multiplier Phototubes. The multiplier phototube (MPT) is the second link in the chain of events leading to scintillation spectrometry (the first is the vial and its sample). We have tested the following tubes: RCA-4501V3 and V4 and EMI-9829QB, all quartz faced, choosing to consider only (i) their response to sample + background from ^{14}C and

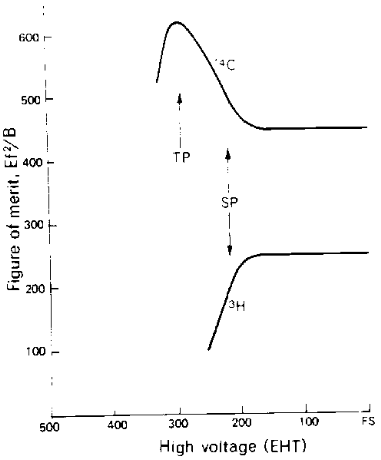

Figure 3. Reduction of EHT applied to MPTs from factory set (FS) value results in a significant increase in FM (Ef^2/B) for ^{14}C and no change in ^3H up to saturation point (SP).

^3H initiated scintillation events (ii) dark current noise and (iii) background in relation to applied high voltage (EHT). The efficiency of the MPTs as a function of high voltage is given in Fig 2. An RCA-4501V4 in a Beckman-7500 LS counter modified to allow precise changes in EHT was used for this test. The % efficiency (Ef) was calculated using unquenched toluene based standards of ^{14}C and ^3H, of ca. 10^5 DPM. The factory set (FS) EHT = 1515V, gave an Ef(^{14}C) = 96% and ^3H = 65%. The effect of reduction of EHT on isotope countrate efficiency was monitored using a lower limit discriminator set at zero (LL=O) and the upper limit (UL) discriminator set at the isotope end-point. As the voltage was reduced, the gain was increased so that maximum efficiency was recorded for each EHT setting. The precision of the plot is high (± 0.1%) and so is the reproducibility. Both isotopes increase in efficiency with increase in EHT. The 'plateau' reached is characteristic for each MPT pair but its length and slope will vary from pair to pair. Thus each MPT pair will have different triggering EHT, slope, length of plateau, EHT value required for maximum efficiency and dark current noise. The dark current noise of this pair of MPTs is also plotted as singles, front (F) and rear (R)

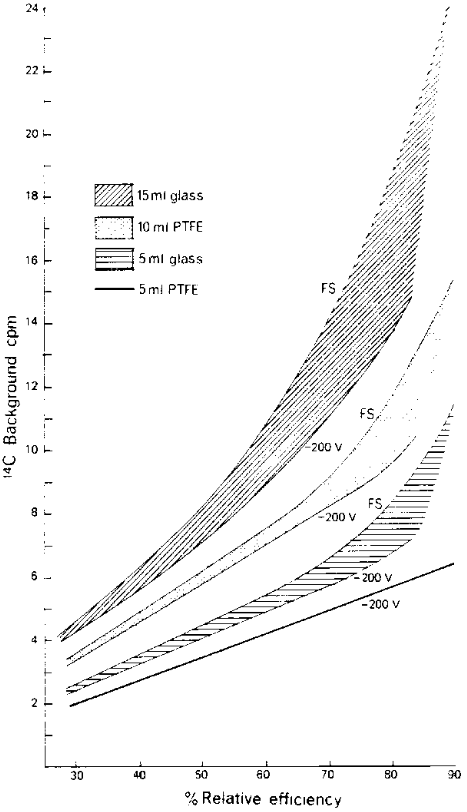

Figure 4. Background (^{14}C) as a function of window width of different efficiencies (%) at factory set (FS) EHT and FS-200 Volts. Vials tested are 15 mL and 5 mL glass and 10 and 5 mL Teflon. All determinations were carried out at balance point.

and sum of F + R. Whilst doing the experiment, we were observing the spectra on a multi-channel analyzer (MCA) and noted that decrease in EHT did not shift the end-point of ^{14}C until a reduction of 215V was achieved. Below this 'saturation point' (SP), the end-point reduced linearily and only an increase in amplifier gain could restore it to its original value. The ^{3}H spectrum did not behave in this manner. The inset in Fig 2 summarizes these findings. We also note that a significant increase in dark current noise (Fig 2) occurs if the EHT is greater than the SP value of 1300 V. Not all MPTs tested gave such a remarkably low dark current noise. Most good tubes have singles values of 10K cpm whilst some would reach 60K cpm. These latter we would reject for low-level counting. The significance of test point (TP) about 32% Ef for ^{3}H (half ^{3}H point) is best demonstrated by changes in FM (EF^2/B) which occur with reduction of EHT (Fig 3). For ^{14}C and ^{3}H, the relationship between background (B) and efficiency (Ef), expressed by FM, remains constant up to saturation point (SP). Then the ^{3}H FM drops sharply whilst the ^{14}C FM increases very significantly (35-40%). For the RCA-4501V4 tested, this corresponds to the inflection test point TP (cf. Fig 2) which, as a rule of thumb, approximates the half ^{3}H point (about 32% Ef).

Background. Background is the countrate observed when counting a sample not containing any radionuclides. For testing the relationship of backgrounds for vials of various sizes and materials - at different EHT applied to MPTs - we have selected a manufacturer supplied background vial containing 15 mL unquenched toluene, a 10mL ANU PTFE vial, a 5 mL masked glass vial and a 5 mL PTFE vial. For a description of these vials see Polach et al[3]. The background (^{14}C), as a function of window width of different efficiencies (%) at factory set (FS) EHT and FS-200 Volts, is illustrated in Fig 4. The efficiency illustrated in Fig 4 was determined by a ^{14}C labelled standard. It is important to note that background (i) is volume related (ii) is window-width related within a given vial size (iii) is significantly reduced, for any vial at any efficiency by reduction in EHT (iv) increases rapidly and non-linearily at factory set EHT efficiencies (Ef > 65%) whilst at a reduced voltage (-200V) the background remains almost linear up to Ef of about 80% for large volumes and truly linear for small volumes and (v) is significantly better for PTFE 5 mL than glass 5 mL, both at -200V. This further confirms the work of Polach et al[3].

To better understand the relationship of background (B) and EHT, we have further determined the B value for a fixed efficiency window (Ef = 80%) both for glass with 15 mL sample and an empty glass vial (Fig 5). Figs 1, 3 and 5 are drawn to the same scale and can be superimposed and in doing so, it will be seen that (i) a saturation effect occurs at or close to the point (SP), and (ii) the background for any vial system is significantly reduced at EHT corresponding to

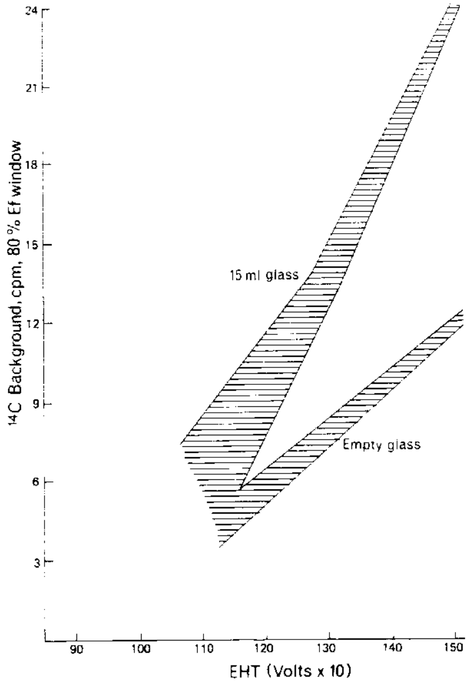

Figure 5. Background (^{14}C) at ~ 80% efficiency window as function of EHT for 15 mL sample in glass and empty glass vial. Fig. 2 and 5 are drawn to the same scale and can be superimposed.

TP = 0.5 ^{3}H without significant loss of ^{14}C efficiency the maximum value of which is 95% at FS and 92% at TP. It is true that the background reduces significantly with further reduction of EHT (EHT below TP). So does the efficiency, the FM (cf. Fig 3), and so will the countrate stability of the system. Thus, nothing is gained by lowering further the EHT. This is true data for what we consider to be an ideal pair of PMTs for low-level counting.

FACTORS AFFECTING PERFORMANCE

Stability. All manufacturers of modern LS counters incorporate electronic features and components which aim to increase the long term stability of their product. As it is not our purpose to make fine distinctions, we consider that all counters on the market today are inherently stable. Many relevant papers have been published, and manufacturer instructions recommend optimal settings for a specific isotope or group of isotopes. We have established that the majority of low-level researchers modify the equipment supplied to them (Table 1) and have further confirmed that reduction in high voltage improves performance for ^{14}C. We shall now examine other, user accessible, parameters which will lead to improvements in signal to noise ratio, whilst maintaining or further improving the stability of LS counters. <u>Gain</u>. The gain of microprocessor controlled counters as well as the high voltage applied to MPTs is set by the manufacturer, often in non user-accessible hardware programs. Older counters have manual controls for some of these functions and all counters have internally accessible electronics which a competent person can and, as Table 1 indicates, has adjusted. Where such adjustments were necessary (i.e. our previously documented reduction in EHT) we recommend that the screw driver adjustable trimpots (as supplied) be replaced with dial calibrated low-noise components. These then will enable changes to EHT gain and coincidence bias (see later) to be made and the effect of such changes to be monitored and, if necessary, will enable the

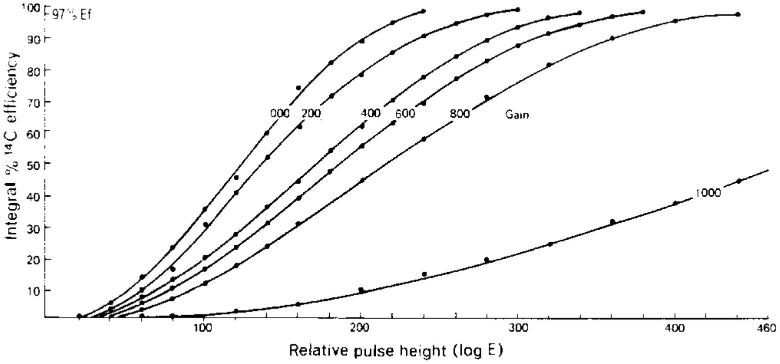

Figure 6a Integral spectra (^{14}C-labelled), log energy (E) of relative pulse height (RPH) at factory set (FS) EHT (1515V, Fig. 1) and variable gain. RPH units represent UL discriminator settings which in 1000 steps cover 0-2 MeV E range. Note stability between Gain = 400-600.

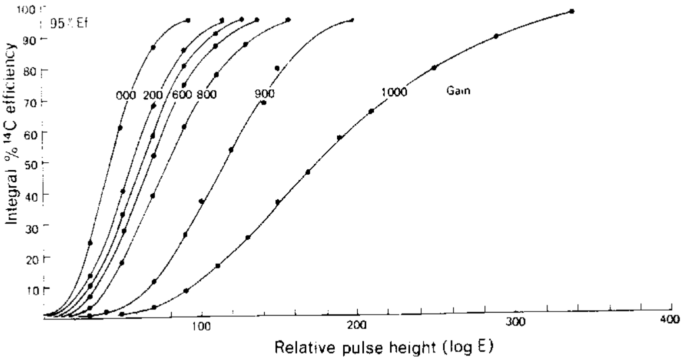

Figure 6b Integral spectra (^{14}C-labelled), log E at FS-215V (1300V = SP of Fig 2) and variable gain. Note increase in stability range between Gain = 200-600.

equipment to be reset to original specifications.

Fig 6a and 6b show that the gain of a counter is not necessarily linear. This statement is based on changes to integral spectra where % efficiency of ^{14}C detection is related to changes in gain and logE (energy) of relative pulse height (RPH). Units are arbitary, 0 and 1000 represent the minima and maxima of the range (0-2MeV). Fig 6a is a plot at factory set (FS) EHT and in Fig 6b EHT is FS-315V (= TP of Fig 2). We conclude that for maximum stability at 1515V (FS) the gain should be between 400 and 600 whilst at 1300V (TP) it can be within the extended range of 200 to 600.

We wish to point out that the background obtained from windows of equal efficiency, is the same at any gain setting for a fixed EHT. In theory this must be so since we are looking at the same scintillation events. In practice, we have confirmed this by many experiments carried out on many types of linear or logarithmic output counters. We think that it is not necessary to supply our considerable experimental evidence as proof that theory and practice agree.

Coincidence resolution time. All liquid scintillation counters, using two 180° opposed MPTs, record pulses initiated within each MPT only if they fall within a set coincidence resolution time interval. The coincidence resolution time, on average 25-40 nano seconds (ns), reduces the probability of accidental coincidence contributing to background counts (B). B is dependent both on random countrate of each tube (N1 and N2) and coincidence resolution time (t) according to $B = 2t(N1)(N2)$. At 10k cpm for each MPT and 25 ns (t) b = <0.1 cpm and at 30k cpm for each and 30 ns (t) accidental B is >1 cpm. Only the latter is significant for low-level counting and the former for ultra low-level counting with which we are at present not concerned. We conclude that selection of MPTs which have an inherent low dark current noise, reduction of EHT below saturation point for ^{14}C (cf. Fig 1) and reduction of coincidence resolution time (t) below 20 ns will each contribute to reduction of background. The sum of these also causes a reduction in efficiency and this overall effect needs to be evaluated in terms of relative Factor of Merit for low-level counting[1].

Coincidence bias. Let us first consider reasons for the function and effect on low-level counting of pulse summation. The pulse summation circuit, common in most commercially available instruments, adds together pulses from the output of each MPT which occur within the specified coincidence resolution time. This results in an improvement in resolution of different energy events; better separation between ^{3}H and ^{14}C isotopes for example. Pulse summation also leads to an increase in counting efficiency as the sum of pulses (sum of pulse height of coincidence events) is likely to be above the factory set coincidence bias threshold. This same effect results also, in an increase in background, as described by Butterfield and Polach[4]. This, from our point of view, is undesirable. Only two manufacturers (to our knowledge) have considered this effect and have minimized it. Searle Analytic use 'lesser pulse' analysis, Laney[5] and LKB-Wallac use a pulse height comparator (an amplitude disparity discrimination system, Soini[6]). Both have the desired effect of reducing background due to optical cross-talk which must not be confused with random coincidence due to dark current noise or afterpulse effects.

We have studied the effect of an increase in coincidence bias on efficiency of ^{3}H and ^{14}C detection on equipment with summed coincidence and a resolution time of 35 and 18 ns. The factory set bias was 3 mV. We were able to extend this range from 1 to 11 mV, reaching the limits of the circuits at these extremes. EHT was varied from factory set voltage (FSV) in steps of FSV -225V, -375V, -465V, etc., as indicated in Fig 7. ^{3}H efficiency was reduced almost immediately the coincidence bias was increased or EHT reduced from factory settings. The ^{14}C efficiency changed progressively allowing a wide range of variations. Based on the radiocarbon dating factor of merit[1], the desirable efficiency range lies within 75 to 85% (shadowed area, Fig 7). It was concluded that an increase in coincidence bias just below the limit of the desired detection efficiency for ^{3}H and ^{14}C (not equivalent to raising the user-accessible lower limit discriminator, LL window setting) will result in a reduction in background, and hence an improved signal to noise ratio. This is due to rejection of lesser pulses initiated by cross-talk of the MPTs. Our improved results, when applying all reduction concepts enumerated above, Table 2, are given later in the text.

Afterpulse decay. Positive ions are produced in the residual gas or at the dynode surface as a result of bombardment by electrons within the photomultiplier. The positive ion is attracted towards the cathode, and if it strikes it with sufficient kinetic energy, a secondary electron pulse will be generated. This pulse will be amplified and will produce a spurious count adding to the background. If the pulse is a result of a previous scintillation, it is called an afterpulse[7]. The total number of afterpulses is linearly related to the light intensity (i.e. number of scintillation photons).

Afterpulses are present in all bi-alkali MPTs, are not dependent on applied EHT level and have a duration time of up to 300 μs[8]. An afterpulse spectrum[9] is shown in Fig 1 and 2. Thermionic emission (dark current noise) will also cause production of positive ions. We have shown that reduction in EHT will reduce the dark current noise to insignificant levels (cf. Fig 2). We suggest that a dead time of 4 ms, following the detection of a scintillation event, will eliminate afterpulse contributions to background under a ^{3}H window. The delay itself, with live-timing, does not affect the performance of LS counters. Afterpulses are below the energy range of the ^{14}C window.

Masking of multiplier phototubes. It is well known that incident

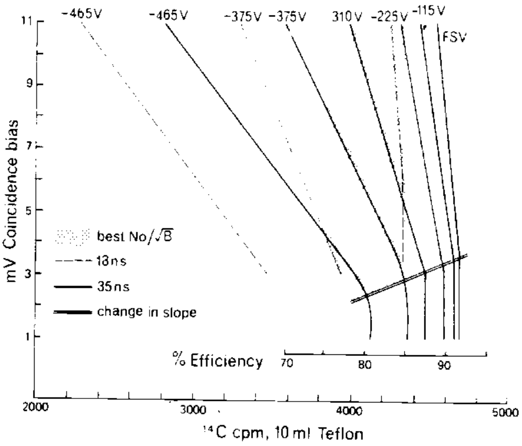

Figure 7. Effect of increase in coincidence bias (mV) on counting efficiency of labelled ^{14}C at various voltages and coincidence resolution times of 35 and 18 ns. Region of merit for low-level ^{14}C is shaded.

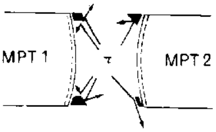

Figure 8. Shape of cross-talk suppression mask and its effect on loss of photons.

ionizing radiations (electrons, neutrons, muons, gammas, etc.) will result in interactions with molecules of the material they impinge on or pass through. Some of these interactions will result in emissions of photons which are added to the true photon count of the radioisotopes we wish to detect. This effect contributes a significant portion of the observed background. Here we are concerned with the background which is observed when there is no counting vial in the counting chamber. This count, as Table 2 indicates, is very significantly higher than the predicted dark current noise coincident events which, for selected low-level MPTs at full voltage, we previously calculated to be <0.1 cpm and which at reduced high voltage

Table 2. Relative performance of LS counters for ^{14}C, each set for highest achievable factor of merit.

Counter No. and specifications[1]	Efficiency %	^{14}C Std (No) cpm	Background cpm	Signal/Noise No/B	Factor of Merit No/ B	Factor of Merit E /B
1: RV,MG	68.5	37.6	6.6	5.7	14.6	711
: RV,MG	81.3	44.6	8.2	5.4	15.6	806
2: RV,M + 5PbG	75.8	41.6	6.8	6.1	16.0	845
3: RV,MG	80.9	44.4	7.8	5.7	15.9	839
: RV,MT	84.4	46.3	4.9	9.4	20.9	1454
: RV,M + 10PbG	73.8	40.5	4.7	8.6	18.7	1159
4: HVG	85.4	46.9	12.6	3.7	13.2	579
: RVG	64.3	35.3	4.5	7.8	16.6	919
: RVT	67.0	36.8	3.3	11.2	20.3	1360
: RVT + 2	67.0	51.5	4.1	12.6	25.4	1095
5: HVG	83.1	45.6	12.0	3.8	13.2	575
: RVG	79.0	43.4	6.7	6.5	16.8	931
: RV,KG	73.6	40.4	4.6	8.8	18.8	1178
: RV,KT	81.6	44.9	3.8	11.8	23.0	1752
: RV,KT + 2	81.6	62.7	4.3	14.6	30.2	1549
: RV,KT - 2	81.6	26.9	2.1	12.8	16.6	3170

1) Specifications: year of manufacture 1965 (1), 1972 (2), 1976 (3), 1980 (4,5); HV = Factory set EHT; RV = Reduced Voltage by 300V; M = 5 mm Mask on MPTs' +5 and +10 = cm of extra Pb all round above factory supplied which in all cases = 5 cm, except LS5 = 3 cm; G = Masked glass vial; T = (Teflon) PTFE Copper Wallac vial; all sample benzene volumes are 5 mL + b PBD except T+2 = 7 mL; T=2 = 3 mL vial; C STD = 95% activity of NBS Oxalic Standard.

(Test Point, Fig 2) becomes insignificant. This empty chamber coincident countrate is commonly called optical cross-talk. A scintillation event, initiating photons within one MPT (face or envelope body), will be seen by the other MPT. If these events are within the coincidence resolution time and, if due to pulse summation, their total pulse height is above the LL discriminator setting, then they contribute to the empty chamber count. Much of the optical cross-talk is enhanced by a light-piping effect of the walls of the MPTs. Some researchers have resorted to placing masks on the edges of the PMTs[10], some suggest changes in reflector configuration,[10,11,12], and some propose the use of Teflon vials as a means of reducing background due to optical cross-talk[1,3,13].

Placing a mask along the edges of the MPTs is a user accessible and easy way to somewhat reduce background. The mask also reduces the efficiency of isotope detection partially due to decreased MPT area and partially due to loss of photons which are reflected out of the system. Fig 8 graphically illustrates the effect different mask designs will have on photon loss.

SOURCES OF INTERFERENCE

Reduction of background and retention of stability of countrates are very desirable features in low-level counting and the results in Table 2 will demonstrate that hitherto suggested electronic changes are successful in achieving both. Reduction in background has, however, one undesirable side effect. It increases the sensitivity of equipment to external sources of interference which are known to contribute non-random or sporadic counts. To demonstrate, let us assume that an unmodified counter has a background, B = 10 cpm, which on 1000 Min count gives an error of 0.1 cpm and an improved counter has B = 3.6 cpm ± .06 cpm (1000 min count). Interference resulting in an injection of 0.1 cpm is within ± 1σ error of the unmodified counter and may not be detected (or be significant) whilst in the improved counter it is almost twice the error and hence has a better chance of interfering. We recognize three main sources of interference.

Line noise. High voltage transients and line transmitted switching noises are known to contribute sporadic pulses. At ANU where high voltage transients are common, we isolate all our counters and computers with line noise suppression transformers. These are available from TOPAZ Electronic Div., USA ('Ultra Isolaters') or ELGAR Corp., USA ('High Isolation Transformers'). They do protect the equipment from decaying oscillatory transients, voltage spikes and under or over-voltages and outages which are the most common modes of power line induced disturbances. For the ANU RC Laboratory, we have selected the 20×10^6 to 1 (146dB) at 5×10^{-4} pF coupling capacitance triple insulated transformers.

Radiofrequency noise. Switches, motors, relays and fluorescent lights generate radiofrequency (RF) interference which is most likely to be picked up by low-level counter circuits (air or line transmitted). Modern, fast counters appear to be more sensitive than their predecessors. Two of our counters (Beckman LS-7500 and Wallac-1215 Rackbeta) have been fitted with an RF pickup whose amplified signal feeds into the coincidence inhibit line. This proved to be a worthwhile arrangement which enables also an additional option, namely an external cosmic ray detector to be fitted above the shield of the liquid counter in an 'umbrella' like fashion. As cosmic particles (muons) are a major contributor to background, their detection and elimination through the coincidence inhibit RF input would substantially improve the performance of any counter. This is a well known anticoincidence cosmic count suppression principle in low-level gas counting which was first described and applied by Anderson, Arnold and Libby[14] to improve detection of environmental radiocarbon. We have not applied it ourselves to LS counting, (however, others have[2]), but foreshadow that we wish to examine the potential of this option.

Static induced noise. Movement of vials in the conveyor belt or cassettes and placement of counting vials into the counting chamber (lift up and down) generates friction which can result in static-electricity build-up on vial surfaces. Static can contribute hundreds of counts. We are, of course, concerned with 1/100th of a count. Known static suppressors are α-particle source emitters which, because of their radiation hazard are not very popular and are prohibited in some countries. Attention to earthing and a time delay, after the vial is in position and prior to commencing a count, eliminate the grossest of interference. For low-level counting, and especially when using plastic vials (which includes Teflon), Wallac Oy, Turku, Finland, have incorporated a high voltage ion source (in all LKB-Wallac beta counters) which discharges the static of a vial at the moment it is entering the counting chamber by ionizing the air around the vial. Both positive and negative static charges are thus effectively discharged. The description of the Wallac HV ionizer is in preparation and will be given elsewhere.

EXPERIMENTAL RESULTS AND CONCLUSIONS

Based on our experience and need for a readily obtainable counter with improved low-level counting characteristics, we have set out to modify an LKB-Wallac 1215 Rackbeta. The Wallac research team provided the specifications and the changes were incorporated by their factory trained engineer on loan from the Australian distributor. The modifications include:

(i) - Phototubes selected for their inherent low background. A certificate (graph) of performance is supplied.

(ii) - switch selectable EHT which allows optium voltage settings for ^{3}H (close to maximum) and ^{14}C, below ^{14}C saturation point (Fig 2) with automatic gain compensation for optimal isotope spectral separation and stability.

(iii) - switch selectable coincidence bias. For ^{3}H at ca. 5 mV and ^{14}C at ca. 80-120 mV to reduce optical cross-talk.

(iv) - switch selectable pulse height ratio comparator. For ^{3}H at 3.8:1 and ^{14}C at 2:1 relative pulse height ratio acceptance levels.

(v) - reduction of coincidence resolution time to <20 ns.

(vi) - an RF frequency amplifier and coincidence inhibit with BNC connection for external (user supplied) cosmic active 'umbrella' shield.

(vii) - a multichannel analyzer interface, enabling accumulation of pulse height spectra for setting up, instruction or data analysis as demonstrated by the windowless data acquisition[9].

(viii) - increased and asymetric shield for external standard to eliminate ES source contribution to background.

(ix) - optional Teflon-copper shielded vials[3].

(x) - optional (not yet available) sealed low-level standards of ca. 200% Modern (2 x NBS Oxalic ^{14}C Reference Std activity) using b-PBD as scintillator and if requests warrant it, low-level ^{3}H standards.

(xi) - line noise isolating transformers (not available from Wallac). If required they should be purchased from suitable manufacturers or their agents.

Experimental results obtained at ANU are given in Table 2. Tests are comparative using counters all of which were optimized for low-level performance by EHT reduction and masking of PMTs or additional lead shielding. The counters are identified as LS1, 2...etc. only, without manufacturers' names. The reason for this is that we do believe that each of them would have responded to the modifications, described above with improved signal to background ratio performance. Thus, the only point we wish to make is that the Kangaroo

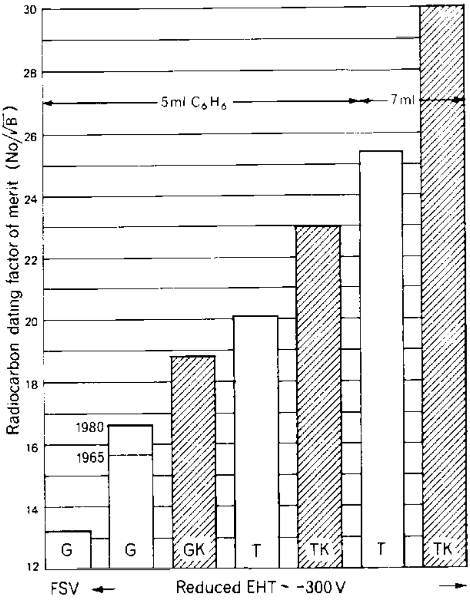

Figure 9. Relative improvements in performance of LS counters for radiocarbon dating. Factory set EHT (FSV) and glass vials, are compared to reduced EHT (-300V), Teflon-copper (Wallac) vials, 5 and 7 mL, and the Kangaroo specifications (K).

modifications, the nickname given to the improved counter incorporating all features described above, do work (Fig 9) and that a Kangaroo model incorporating these features is now available through the LKB-Wallac distributors.

The improvements do not place the Kangaroo into high precision ultra-low level category. To do this, an active cosmic guard and additional lead shielding are required. However, the Kangaroo will enable researchers to enter the low-level counting field, routinely, without the need to resort to 'home-made' modifications, as was the case until now.

ACKNOWLEDGEMENTS

We wish to thank all researchers who have supplied us with data and details of their procedures and Linbrook International, the Australian LKB-Wallac distributor for support throughout this project. John Gower, ANU RC Lab., has carried out many of the tests described. We wish him a happy retirement. Winifred Mumford, Canberra, prepared the art work with great skills and Maureen Powell, ANU RC Lab., carefully typed the manuscript. We thank them both.

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