[RADIOCARBON, VOL 29, No. 1, 1987, P 12–17]

SCINTILLATION COUNTER PERFORMANCE AT THE SMU RADIOCARBON LABORATORY*

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ABSTRACT. Results are presented of a study of counter performance and vial characteristics for three liquid scintillation counters used at the SMU Radiocarbon Laboratory: the Intertechnique LS20, Packard Tri-Carb 460C, and LKB Wallac Rack Beta 1217. Modifications to photomultiplier tube high voltage, pre-amplifier gain, energy window settings, counting vial design, and sample holder design have resulted in reduced background, higher counting efficiency, and greater long-term stability for the Intertechnique and Packard counters. Square quartz counting vials are used in the Intertechnique and Packard counters with excellent results. Use of Teflon vials in the LKB counter requires careful cleaning procedures and long counting times.

COUNTER AND VIAL CHARACTERISTICS

The SMU Radiocarbon Laboratory uses three liquid scintillation counters: an Intertechnique LS20, a Packard Tri-Carb 460C, and the low-background "Kangaroo" version of the LKB Rack Beta 1217. The Intertechnique is a benchtop non-cooled model with the following characteristics:

1) carefully selected photomultiplier (PM) tubes (RCA 4501V3) with low background and high counting efficiency at reduced dynode voltage,

2) a shielded PM tube assembly made from low-activity lead,

3) a modified single-sample manual loading drawer that accommodates quartz vials holding 3, 1.5, 0.6, and 0.3ml counting solutions (Haas, 1979).

Temperature sensitivity of counter components requires careful control of laboratory environment. It has been observed that background count rates increase 0.1-0.4cpm during high-temperature excursions, as timing oscillator and energy window settings are temperature dependent. The counter has enjoyed long-term stability when laboratory temperature has been kept stable (± 1°C). The counter has been modified by removal of the ^{137}Cs standard from the cabinet and by replacement of variable potentiometers by fixed resistors for energy window adjustments.

The Packard counter is a floor-standing model with built-in cooling. Cooling is disabled, however, to avoid moisture condensation within the counter. Sample loading is automated, and 15 programs can be stored for different 10-sample trays. At present, a separate program (tray) is used for each of 4 counting vials (3 at 3ml and 1 at 1.5ml filling volume). Several modifications were made to this counter:

1) Following a one-year trial period, the vial lift and shutter mechanisms were modified to accept square quartz vials. These vials are inserted in a round black nylon holder with the same outside dimensions as a standard 15ml vial. In the cylindrical wall of the holder are two opposed windows through which PM tubes view the vial. Proper alignment of the holder

* This paper is a modified version of the one presented at the Twelfth International Radiocarbon Conference in Trondheim, Norway, June 24-28, 1985.

is maintained with a slot along its outside and several guide pins in the lift tube and sample chamber.

2) The original analog amplifier board was replaced by Packard Instruments with a design having greater stability and adjustable gain. Gain was boosted to spread the energy spectrum and to separate the background and ^{14}C peaks for energy window adjustment. PM tube high voltage (HV) was decreased to the lower edge of the weakly developed plateau of the PM tube HV *vs* counting efficiency curve (Fig 1).

3) A focusing and masking insert (similar to the design of Butterfield & Polach, 1983) was placed before each PM tube to reduce crosstalk and reflect scintillation photons to the center of the PM tube front dynode.

4) The ^{226}Ra external quench standard was removed since the channel ratio method, rather than external standardization, is used for quench determination. External standardization is not used due to possible PM tube and counting solution memory effects following standard-generated counts, and to questions regarding the effectiveness of internal shielding against the ^{226}Ra standard.

5) Frosted quartz vials replaced clear vials, with resultant background decrease (Table 1) due to crosstalk reduction.

Improvements in counter performance with each modification are demonstrated by increased counter efficiency and decreased background (Table 1). Counting solutions comprise benzene with 0.9100 weight percent butyl-PBD scintillant.

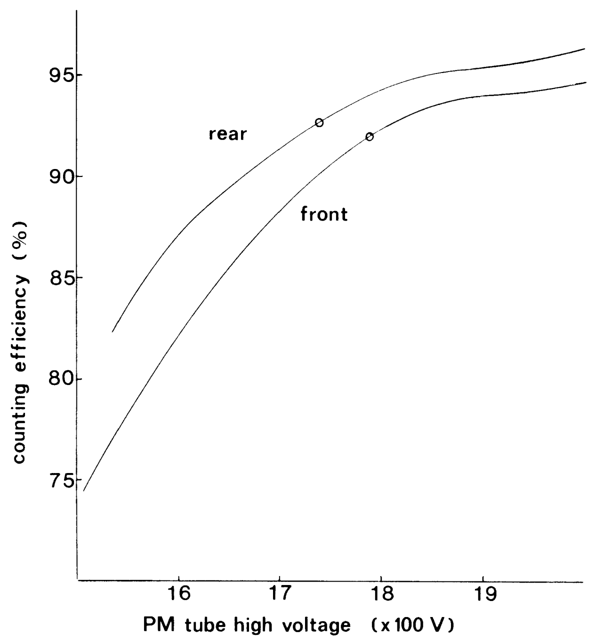

Fig 1. Efficiency *vs* PM tube HV for Packard 460C. HV settings for front and rear PM tubes = (0).

TABLE 1
Counter and vial response characteristics

Counter	Vial (ml)		Background (cpm)	Efficiency (%)	E^2/B*	Nox/√B**
Intertechnique LS20	3.0		2.6	67.8	1768	13.8
RCA 4501V3 PM tubes	1.5		2.2	68.6	2139	7.3
Clear square quartz vials	0.6		2.1	70.1	2337	3.2
	0.3		1.9	68.6	2477	1.7
LKB Rack Beta 1217 Copper/Teflon vials CBC option	3.0		1.8	71.6	2850	17.4
	7.0		3.8	80.8	1706	32.9
Packard Tri-Carb 460C RCA 4501V3 PM tubes						
1. Initial testing	15.0		4.4	76.9	1344	12.0
Cylindrical glass vials	15.0 masked to 3ml vol		4.9	80.0	1306	5.3
2. First upgrade Clear square quartz vials, flat reflectors	3.0	A ch	5.9	83.6	1185	11.3
		B ch	2.0	52.9	1400	12.2
	1.5	A ch	5.0	83.0	1380	5.9
		B ch	1.4	50.9	1850	7.0
3. Present setup Frosted quartz vials in black nylon holders, curved reflectors	3.0	A ch	5.4	82.7	1270	11.6
		B ch	2.7	64.7	1550	12.4
	1.5	A ch	4.7	80.8	1390	5.9
		B ch	2.0	58.0	1680	6.7

* E^2/B Figure of merit = (Counting efficiency^2)/Background cpm
** Nox/√B Radiocarbon Factor of merit = (0.95 Mod std cpm/√Background cpm)

The LKB Rack Beta 1217 counter (low-background "Kangaroo" version), a floor-standing non-cooled model, had acceptable performance characteristics as delivered; however, counting channels were reset from default values to windows allowing maximum ^{14}C detection efficiency using Teflon vials. The LKB Teflon/copper 3ml counting vials required redesign to promote easier sample weighing and reduced sample loss. The original vial design was changed with the addition of a lightweight Teflon screw cap to enable weighing on our analytical balance. The seal between the Teflon cap and vial is maintained by a Teflon disk backed by silicone. Tests have shown that the Teflon vial/cap assembly absorbs on occasion ca 0.5mg over a two-day counting period, presumably due to laboratory humidity fluctuations. Observations of vial memory during alternating runs of background and modern standards (0.5cpm background increase following modern standard runs) resulted in our adoption of a cleaning procedure more extensive than that recommended by the manufacturer; vials are rinsed four times in benzene, and vacuum-dried at 60°C for two hours. Vials are then placed in a desiccator until needed for counting, and allowed to equilibrate one hour before filling. Filled vials rest in the counter for at least 1.5 days before counting begins because of their high sensitivity to light, and require a much longer time to stabilize at the start of counting than quartz vials. The first five 100-minute counts are routinely omitted to remove effects of initial erratic data. Given these constraints, Teflon vials provide low background and high counting efficiency and figure of merit (Table 1; Gupta & Polach, 1983).

ENERGY WINDOW SELECTION

Energy window adjustment is a necessary procedure in preparation of a counter for routine operation. The energy distribution of background and ^{14}C pulses differs for each counter and different considerations apply in the choice of lower and upper limits for ^{14}C counting. Spectrum measurements are made easily in the Packard counter, which allows immediate access to any stored channel or group of channels, enabling listing of a complete spectrum. Spectrum measurement in the Intertechnique and LKB counters is time consuming, however, as data must be collected for each channel or group of channels before proceeding to the next set of channels. It has been demonstrated that use of an external multichannel analyzer (Polach *et al,* 1983b) hastens this procedure appreciably.

The Intertechnique counter has a logarithmic amplification stage and presents a nearly symmetric ^{14}C spectrum (Fig 2A). The background curve is nearly flat; thus, window settings are determined wholly by the ^{14}C curve. A wide window records counts in the entire spectrum; setting of a narrow window on the steep high energy flank of the curve acts as a sensitive quench indicator (Baillie, 1960; Gupta & Polach, 1983). A curve shift to lower energy due to quenching causes a proportionally larger drop in measured output in this narrow channel compared to energy loss in the wide window setting. Thus, the two-channel ratio method makes application of an external radioactive source unnecessary.

The ^{14}C curve of the LKB counter (Fig 2B) is similar to that of the Intertechnique counter. The background spectrum is flat in the region of interest and rises steeply slightly beyond endpoint. As in the Intertechnique, two windows are set for quench monitoring. The Wallac amplitude disparity discrimination (CBC) option, based on coincidence timing delay via shaping of PM tube pulses (Soini, 1975), accepts a pulse only if pulse height ratio is $< 2:1$ (for ^{14}C) within the coincidence resolution interval (Polach *et al,* 1983a). Use of CBC halves background count rate and reduces counting efficiency ca 8%. Energy window settings and background cpm are identical for each Teflon vial in use, given strict cleaning and cpm monitoring procedures.

The Packard counter presents linearly amplified spectra compared to logarithmic output of the Intertechnique and LKB counters. Initial tests revealed that the sharp background and ^{14}C peaks overlapped completely (Fig 2C); it was not possible to reduce background without substantial ^{14}C count loss. Increase of pre-amplifier gain and reduction of PM tube HV lowered the background peak and separated, partially, background and ^{14}C peaks; introduction of square quartz vials and changes in the optical geometry of the counting chamber spread the ^{14}C spectrum (Fig 2D). The present arrangement sets the lower A-channel window limit within the wide ^{14}C peak (Fig 2E); the background peak lies slightly below the lower window limit. Some background reduction was achieved by frosting the sides of the square quartz vials, which dampens crosstalk. B-channel lower limit is set so

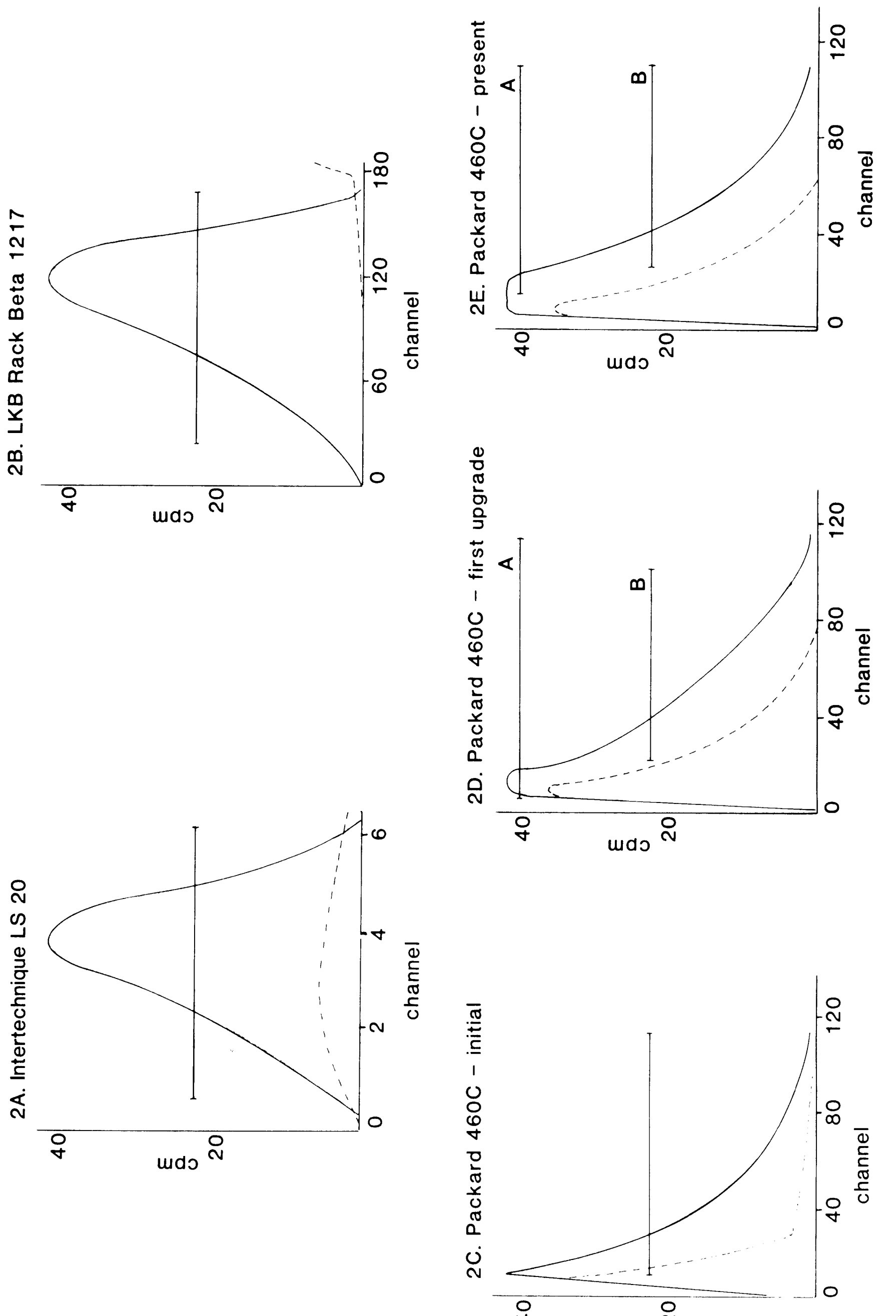

Fig 2. Modern standard (——) and background (---) spectra for counters. Window settings = (|—|).

that the majority of the background peak is distinctly below the lower window limit. Part of the ^{14}C spectrum is excluded, and the upper window limit is positioned near the endpoint of the ^{14}C curve. B-channel counting efficiency is reduced (Table 1), and count rate is sensitive to quenching. A discrepancy in sample age calculated from A and B channels (B-channel age > A-channel age) is, thus, a sensitive quench indicator.

SUMMARY

Several modifications have optimized our three liquid scintillation counters for routine ^{14}C dating. Reduction of PM tube HV to decrease dark current noise coupled with increase in gain to boost counting efficiency, improvements in optical geometry for the use of square quartz vials, use of low background materials for sample holders (lead, black nylon), and energy window adjustment for quench detection have yielded reduced background and greater stability in counter operation.

ACKNOWLEDGMENTS

The support of NSF grant BNS 8211974 is gratefully acknowledged. We thank H Polach and R Kalin for their critical comments.

REFERENCES

Baillie, L A, 1960, Determination of liquid scintillation counting efficiency by pulse height shift: Internatl Jour Appl Radiation Isotopes, v 8, p 1–7.

Butterfield, D and Polach, H, 1983, Effects of vial holder materials and design on low-level C^{14} scintillation counting, *in* McQuarrie, S A, Ediss, C, and Wiebe, L I, eds, Advances in scintillation counting: Edmonton, Univ Alberta Press, p 468–477.

Gupta, S K and Polach, H, Radiocarbon dating practices at ANU: Research School Pacific Studies, ANU, Canberra, 173 p.

Haas, H. 1979, Specific problems with liquid scintillation counting of small benzene volumes and background count estimation, *in* Berger, R and Suess, H E, eds, Radiocarbon dating, Internatl ^{14}C conf, 9th, Proc: Berkeley, Univ California Press, p 246–255.

Polach, H, Nurmi, J, Kojola, H, and Soini, E, 1983a, Electronic optimisation of scintillation counters for detection of low-level H^3 and C^{14}, *in* McQuarrie, S A, Ediss, C, and Wiebe, L A, eds, Advances in scintillation counting: Edmonton, Univ Alberta Press, p 420–441.

Polach, H, Robertson, S, Butterfield, D and Gower, J, 1983b, The "windowless" approach to scintillation counting: low-level C^{14} as an example, *in* McQuarrie, S A, Ediss, C, and Wiebe, L I, eds, Advances in scintillation counting: Edmonton, Univ Alberta Press, p 494–507.

Soini, E, 1975, Rejection of optical crosstalk in photomultiplier tubes in liquid scintillation counters: Wallac rept, Turku, Finland, 9 p.