CHAPTER 43

Liquid Scintillation Counting Performance Using Glass Vials in the Wallac 1220 Quantulus™

Lauri Kaihola

ABSTRACT

Low-potassium glass vials can obtain reduced background count rates for beta particles in liquid scintillation counting with the aid of pulse amplitude comparisons and pulse shape analysis; they also retain relatively high counting efficiencies. Adding secondary fluor to the solvent, to shorten the pulses from sample decay events, helps the effectively separate these from the slow glass fluorescences.

The power of this method in improving counting performance was examined for:

(1) ^{14}C in benzene in normal and low background environments using the Wallac low-level liquid scintillation counter, 1220 Quantulus™, with an active anticoincidence guard detector. The primary fluor was 15 mg/mL butyl-PBD, and the secondary fluor was 1 mg/mL bis-MSB,

(2) ^{3}H in aqueous solution with commercial cocktails.

In a normal environment, using Wheaton 20 mL low-^{40}K vials filled with 5 mL of benzene, the best ^{14}C figures of merit were achieved in a window of 65% counting efficiency. It gave a 0.78 cpm background count rate using low bias with the pulse shape analyzer (PSA) alone and a 0.51 cpm background count rate using the pulse amplitude comparator (PAC) in conjunction with PSA. The corresponding figures in a low background environment were 0.57 and 0.27 cpm.

Tritium background count rates were 2.8 and 2.9 cpm at 27 and 22% counting efficiency using Quickzint™ 400 and Optiphase Hisafe™, respectively, with PSA + PAC.

INTRODUCTION

Using glass vials in low-level liquid scintillation counting offers some advantages over plastic and teflon vials. The glass is inert and easy to clean (cleanliness can be checked by visual inspection). It is dimensionally stable and impermeable to aromatic and volatile solvents such as the benzene used in ^{14}C dating. For long term low-level counting, the accompanying plastic caps can be replaced with metal caps lined with indium gaskets to prevent vapor loss of the volatile sample.[1] Teflon vials were specially designed for counting ^{14}C using benzene and still reach the best performance, but they require very careful calibration and cleaning.[2] The search for an ideal vial material, in terms of low-level counting performance, is in progress; quartz might be the one.[3,4,5] However, the cost of inherently low radioactivity synthetic quartz is very high. The ability to use standard low-^{40}K glass vials would be a good choice for low-level counting if the background can be reduced to acceptable levels at a relatively high counting efficiency.

METHOD

Glass vials exhibit unquenchable and quenchable background radiation components. The unquenchable background is caused by 1.3 MeV beta particles emitted in the ^{40}K decays in the glass and by high energy particles interacting with the vial and PMTs (approximately 10 cpm in the Quantulus™; this is seen with empty glass vials as well). Further, fluorescence events are also created in the glass, mainly in the low energy region of the spectrum. These pulses are slower than the Cerenkov radiation created within the glass. This part of the background severly interferes with ^{3}H beta spectrum. The quenchable, mainly high-energy background, is caused by photons from ^{40}K decays in the vial and PMTs; the result is a Compton continuum in separable from a beta particle spectrum. Beta particles from ^{40}K decays and alpha particles (U,Th) from the inner surface of the vial reach the cocktail, contributing to the high energy background.[6]

Bias Threshold

The Quantulus has a user controlled bias threshold which can be set either low or high. The acceptance levels of the pulse amplitudes are tested for both PMT signals individually, before the coincidence summing of pulse heights.[7] At low bias, maximum ^{3}H detection efficiency is retained, while high bias effectively rejects cross talk, Cerenkov, chemiluminescence, and low energy pulses (e.g., part of the ^{40}K spectrum in glass and most of the ^{3}H spectrum). It is very effective in reducing background for ^{14}C, but it cannot be used for this purpose with low energy radiations.

Pulse Amplitude Comparator

The Wallac Quantulus electronics contain a user adjustable pulse amplitude comparator (PAC) that can to reject those events in the PMT tubes which vary more in their amplitudes than specified by their selected PAC setting.[8,9] PAC is designed to discriminate differently between low energy (^{3}H and others) and high energy (e.g., ^{14}C) pulses; the lower the energy of the ionizing event, the milder the PAC effect. The ^{14}C background cross talk events at low bias are more likely to be rejected by PAC due to their greater pulse amplitude disparity than the sample decay events. In this way one is also able to improve the ^{14}C S/N ratio without significant efficiency losses.

Pulse Shape Analysis

The pulse shape analyzer in the Quantulus is an analog device which integrates the tail of each pulse.[10] This integral is then compared with the total pulse integral to produce an amplitude independent parameter that relates to the pulse shape. Typically a true beta pulse decays exponentially in a few nanoseconds, while an alpha pulse decays non-exponentially in a few hundred nanoseconds. The application of this pulse shape parameter is user selectable and variable. Pulse shape analysis (PSA) is primarily intended for alpha/beta particle spectrum separation. It can also be used for background reduction, provided that the pulse shape of the sample and background signals differ.

Glass Vial Backround Reduction

In ^{14}C counting of benzene, butyl-PBD is widely accepted as the fluor due to its high efficiency and quench resistance in the presence of impurities in the sample solute.[2] It is, however, a quite slow fluor requiring the addition of a secondary fluor, e.g., bis-MSB, to improve pulse shape contrast between the fluorescent glass vial backround events and the fast ^{14}C sample pulses. The secondary fluor shifts the emission peak wavelength. This is essential to matching the peak transmission of the guard in the new Packard low-level counters, where it aids in achieving higher counting efficiency at lower background.[11,12,13]

The scheme was tested in an ultralow background liquid scintillation spectrometer, the Quantulus, inside and outside our low-level laboratory at Wallac. The Quantulus is fitted with a cosmic guard counter whose performance is totally independent of sample characteristics. There cannot be any beta particle energy transfer out of the actual sample spectrum into the guard. Therefore user selectable PSA enable free discrimination of long fluorescent pulses from the fast sample events in the glass, thus reducing the background without losing any efficiency through erroneous interpretation of their origin. Both the accepted and rejected pulse spectra can be analyzed and recorded to evaluate PSA and PAC performance at different settings and under different cocktail and environmental conditions.

RESULTS

Radiocarbon

Standard 20 mL low-^{40}K glass vials (Wheaton) were used for the experiments at low-bias settings. The PSA settings were scanned from 1 to 25 in steps of 5,

Table 1. ^{14}C Efficiency and Background Variations, Using 20 mL Low-^{40}K Glass Vials with a 5 mL Benzene Sample, in a Normal and Low Radiation Environment with PSA and PSA + PAC, at low bias

Mode Efficiency ^{14}C %		PSA B cpm B reduction %		PSA + PAC B cpm B reduction %	
		N	LR	N	LR
77%	B =	1.34	1.00	0.78	0.49
	E^2/B =	4400	5900	7600	12100
	B_{red} =	46%	59%	72%	82%
65%	B =	0.78	0.57	0.51	0.27
	E^2/B =	5400	7400	8450	15650
	B_{red} =	31%	38%	56%	67%

Note: B = background cpm
B_{red} = background reduction % in the selected window with respect to no PSA or PSA + PAC electronic discrimination
N = normal environment
LR = low radiation environment

and the PAC was scanned from 180 to 250 in steps of 10. These setting ranges have perceptible effects on the counting performance in the cocktail, 15 mg/mL butyl-PBD and 1 mg/mL bis-MSB. The sample 5 mL volume was placed in an unmasked vial. Spectroscopic grade benzene was used as the background sample. ^{14}C labeled fatty acid was used as the reference standard. Performance figures are given at optimum windows and at balance point, with the highest figure of merit (E^2/B) for the specified vial type, size, and sample volume (Table 1). Typically, a reduction of 31 to 72% in the background is observed at 65 and 77% efficiency windows when only PSA or PAC and PSA are activated, respectively, in a normal environment. The background is reduced by some 38 to 82% from the original when PAC also is activated in a low radiation environment. There is some 40% reduction in backround count rate when the instrument is taken from a normal environment into the low-level laboratory. More significantly, however, when using PAC + PSA, there is a background reduction in low-^{40}K glass vials of 82% at a counting efficiency of 77%, and a 67% reduction at 65% efficiency.

Tritium

The ^{3}H beta emission in aqueous solutions and glass fluorescence pulse amplitude spectra are very similar and overlap. The highest figure of merit is therefore achieved in the widest window. Water samples were tested as a mixture of 8 mL H_2O and 12 mL scintillation cocktail, Quickszint™ 400 from Zinsser and Optiphase Hisafe™ 3 from Pharmacia-Wallac. In a low-level radiation environment the background count rate in a full window was 2.8 and 2.9 cpm respectively, at 27 and 22% counting efficiencies, using PSA at maximum effect and PAC at 255. In a normal environment the increase is marginal, on the order of 0.5 cpm, leading to background count rates less than 3 cpm at efficiencies up to 26% for aqueous solutions.

In Teflon vials the best reported performance gave a background of 0.42 cpm at 27.9% with Quickszint 400.[14] In a normal environmental gamma flux the background would be below 0.8 cpm. In plastic vials the background is marginally higher (0.05 cpm). Modern biogradeable cocktails do not cause problems with Teflon and plastic vials, therefore there is no need to run low background water samples in glass vials; plastic vials are equally cheap and give better performance. Counting in Teflon and plastic vials in the Quantulus allows direct measurement of most environmental ^{3}H samples without enrichment.

DISCUSSION AND CONCLUSIONS

The use of standard low-^{40}K glass vials is of merit, and it is recommended for low-level ^{14}C determinations when maximum resolution of weak signals (close to background) is not essential. This is the case with close to Modern samples, while for very old samples, Teflon retains its merit due to very high counting efficiencies and ultralow background count rates. Application of PAC and PSA at high bias does not improve the performance of Teflon vials. In glass vials background reductions are achieved at the expense of some 10 to 20% loss in counting efficiency.

Optimum performance is not critical in all applications. Moreover, it depends on environment, vial, sample purity (lack of quench), cocktail selection, and concentration. The inherent stability of the Quantulus ensures reproducible performance under optimal counting parameter settings.

Performance in the ^{3}H energy region is affected by the residual radioactivity of the low-^{40}K glass, hence true, low background counting is not possible. Plastic and Teflon vials make use of the full power of low-level liquid scintillation spectrometers.

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