[RADIOCARBON, VOL 32, NO. 3, 1990, P 381-386]

# TIME-RESOLVED LIQUID SCINTILLATION COUNTING

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ABSTRACT. Historically, scientists who perform low-level measurements of $^{14}C$ for age dating, and $^{3}H_2O$ for environmental contamination, have purchased or constructed highly specialized instruments to quantitate low-level radionuclides using a general-purpose liquid-scintillation analyzer (LSA). The LSA uses special time-resolved 3-D spectrum analysis (TR-LSC) to reduce background without substantially affecting sample counting efficiency. This technique, in combination with a special slow fluor scintillating plastic, further reduces the minimal detectable limit for the TR-LSC liquid scintillation counter.

## INTRODUCTION

To quantitate low-level samples in an LSA, it is important to distinguish true background pulses (environmental and instrumentational) from scintillation pulses produced by a decay of a beta-emitting nuclide. Thus, we investigated the various type backgrounds. There are four types of background that contribute to the total spectrum: the instrument, cross-talk, radioactivity from glass vial and photomultiplier tube (PMT), and scintillator background. The instrument background is the result of electronic noise in the LSC system. The energy spectrum of this background is low and represents 10% of the total observed background. The second background is cross-talk between the two photomultiplier tubes. This background is the result of the spontaneous release of photoelectrons from the cathode of one or both of PMTs. It is low energy (>20 keV) and represents 20% of the total observed background. The third type of background is from the natural radioactivity ($^{40}K$) that occurs in the glass vial and the glass of the PMTs. This background energy has a flat distribution over the entire range out to 2000 keV and represents 40% of the total background. The fourth background is from the cosmic and environmental radiation, which can interact with the scintillation solution. This represents 30% of the total background, and covers a broad energy range (0-2000 keV).

The question is, can any or all of these four backgrounds be eliminated without substantially decreasing the counting efficiency of the sample? Are there some characteristics of these backgrounds that can be used to distinguish the (environmental and instrument) background from true nuclear decay? To characterize the background and beta pulses, the shape and components of both the beta decay and background must be carefully analyzed. The first to be characterized is the scintillation pulse originating from a beta decay.

A typical beta scintillation pulse (Fig 1) consists of a fast/prompt pulse component and may contain a delayed pulse/slow component. The delayed pulse is found in samples that are flame-sealed and purged with argon or nitrogen, which can act as a quenching agent to remove any oxygen from the sample. The fast pulse is sharp with a pulse width of approximately two nano-seconds. The nuclear decay pulses are the result of quenchable pulses that result from the cosmic or environmental radiation interaction with the scintillator or a true beta decay. The background pulses resulting from instrumentation noise, cross-talk, and naturally occurring radionuclides present in the glass or the PMT or the sample vial, are called non-quenchable because they are not affected by the components in the sample or scintillation solution and cannot be quenched. Close examination of non-quenchable background pulses shows substantially different characteristics from those of the beta scintillation pulses. The non-quenchable background results from the interaction of background radiation with the material in the environs of the counting chamber. Figure 2 shows pulses created by non-quenchable backgrounds.

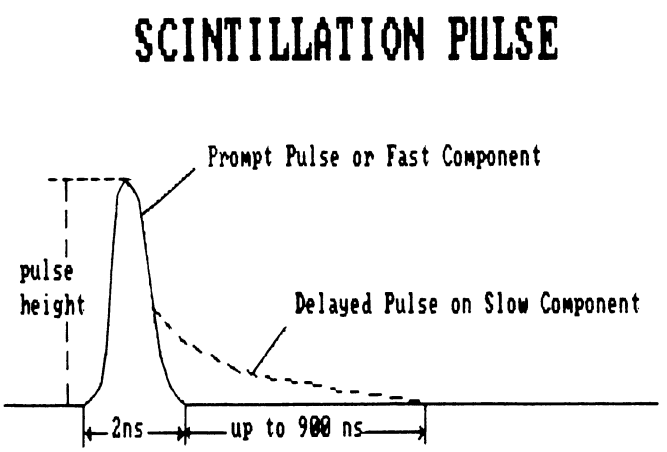


Fig 1. Typical beta scintillation pulse

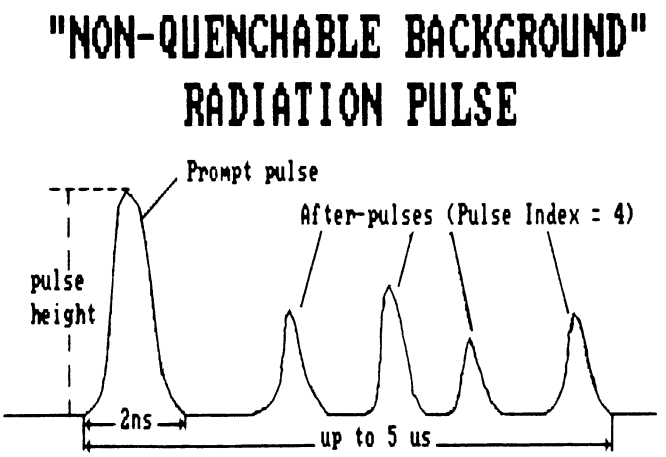


Fig 2. Non-quenchable background pulse

These pulses from non-quenchable background consist of a major pulse (fast component), resembling a beta scintillation five microseconds after the main prompt pulse. The number of low-intensity "afterpulses" can be different from one background pulse to the other; this can be due to low-intensity Cerenkov radiations in the glass envelope of the PMT and surrounding material or natural $^{40}K$ in the glass. The total number of "afterpulses" is called the Pulse Index, and are counted for five microseconds after the main prompt pulse.

The traditional energy plot for a sample is two-dimensional representing count rate as a function of energy. Using the new 3-D afterpulse technique with the pulse index in the third dimension, we obtained a true 3-D plot. Figure 3A shows the 3-D plot for a background spectrum with afterpulses occurring after almost all pulses, up to 15 afterpulses. On the other hand, a $^{14}C$ sample from the benzene synthesizer shows counts in only the zero to three afterpulse spectral planes (Fig 3B).

If the 3-D plot (Fig 3A) sample background is subtracted from the 3-D plot (Fig 3B), $^{14}C$ sample, then only the true beta pulses from the sample can be quantitated. This analysis results in a reduced background without a substantial loss in counting efficiency of the sample.

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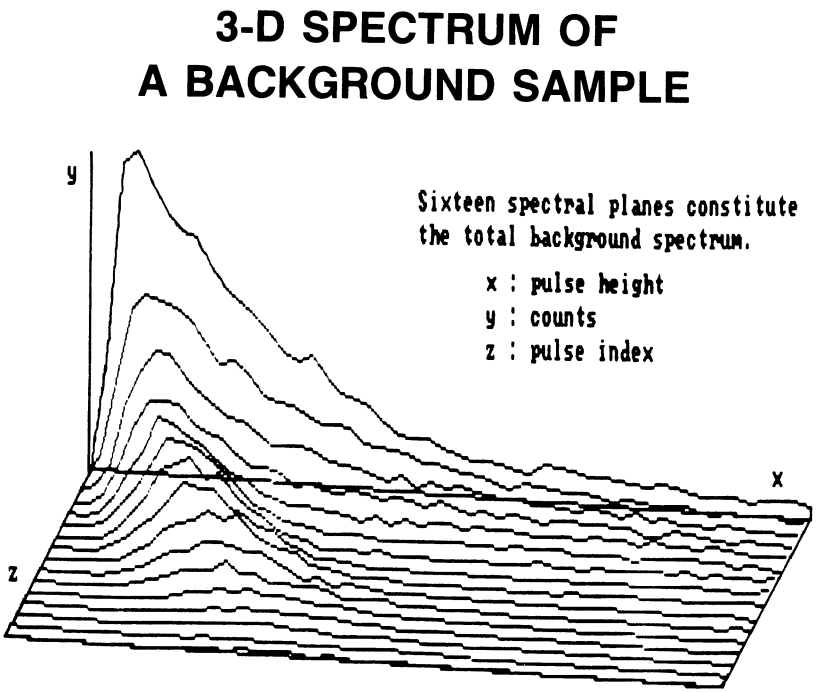
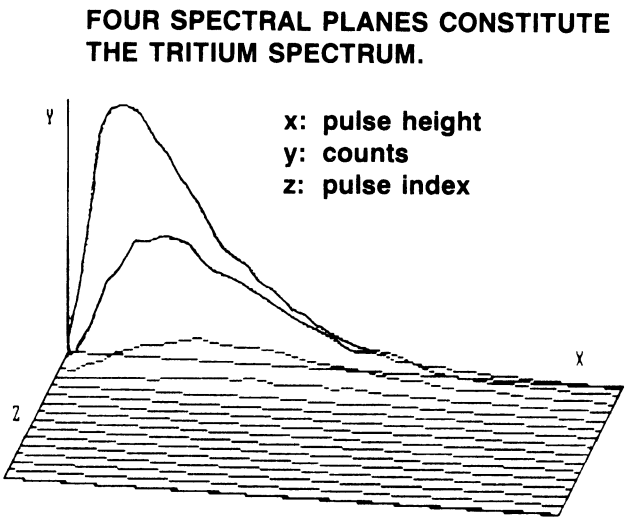



Fig 3A. 3-D spectrum of background sample




**3-D Spectrum of Unquenched $^{3}H$**

Fig 3B. 3-D spectrum of $^{14}C$ sample (benzene synthesizer)

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## RESULT

Based on these characteristic differences between the real beta-decay distribution and the background radiation spectrum we can discriminate from real beta-decay events. Using the pulse index as a discriminator we can exclude background pulses from the $^{3}H$ or $^{14}C$ spectrum. We can obtain $E^2/B$ optimization by selecting the optimal pulse index discriminator setting. Table 1A and B show typical improvement in the $E^2/B$ achieved for both tritium and $^{14}C$, counting the samples for 600 minutes, or 0.5% 2s, whichever occurs first.

TABLE 1

Effect of pulse index discrimination on background, efficiency, $E^2/B$

*A. Tritium*, 0.5-5.0 keV region, $O_2$ quenched (10ml InstaGel and 10ml $H_2O$ in 20ml glass vial)

| Amount of pulse-index distribution | $^{3}H$ efficiency | Background (CPM) | $E^2/B$ |
|---|---|---|---|
| None | 26.50 | 18.45 | 38.06 |
| Normal (minimum) | 26.24 | 12.75 | 54.08 |
| High sensitivity (moderate) | 24.68 | 9.25 | 65.85 |
| Low level (maximum) | 22.59 | 3.33 | 153.25 |
| A 402.6% increase in $E^2/B$ | | | |

*B. $^{14}C$ benzene*, 10-102 keV, $O_2$ quenched (3.5ml benzene scintillator in 7ml glass vial)

| Amount of pulse-index distribution | $^{14}C$ efficiency | Background (CPM) | $E^2/B$ |
|---|---|---|---|
| None | 83.45 | 9.67 | 720.15 |
| Normal (minimum) | 81.87 | 7.07 | 948.05 |
| High sensitivity (moderate) | 78.50 | 4.74 | 1300.05 |
| Low level (maximum) | 70.70 | 1.38 | 3560.00 |
| A 495.5% increase in $E^2/B$ | | | |

The data in Table 1 clearly show much-improved counting sensitivity using this unique patented TR-LSC technique. Counting sensitivities achieved by this technique rival those previously possible only by the use of very large amounts of passive shielding and/or active (anticoincidence) shielding.

After further study of the burst-pulse-counting discrimination method as a beta pulse validation scheme, it was clear that the difference of a few afterpulses immediately following a coincident event could affect the discrimination performance. To further improve the detection capacity of the burst-counting electronics, we had to amplify the number of afterpulses. We determined that a long decay time scintillator would provide the amplified number of afterpulses needed to improve the burst pulse discrimination. Since the purpose was to differentiate background, we decided that a slow fluor scintillator physically external to the sample would act as an active guard shield.

Thus, when a slow fluor external to the sample was excited by a high-energy background (cosmic or environmental radiation), an LSA circuitry to differentiate this background from a true beta decay produced within the scintillation vial. Since the guard was a plastic scintillator material, a slow component was produced with several afterpulses. This plastic or glass scintillator was formed into a vial holder or a detector guard that filled the area between the scintillation vial and the photomultiplier tubes. Tables 2 and 3A/3B show the results for $^{14}C$ and $^{3}H$ using these special plastic scintillators (vial holder and/or guard detector).

TABLE 2
$^{14}C$ benzene in PPO (5gm/L) and POPOP (0.2gm/L) (optimized regions)

| Amount of pulse index | Sample | $^{14}C$ efficiency | Background | $E^2/B$ |
|---|---|---|---|---|
| None (3-D) | Sample only | 64.99 | 3.62 | 1167 |
| Maximum | Sample only | 54.04 | 0.76 | 3844 |
| Maximum | +vial holder | 60.99 | 0.63 | 5904 |
| Maximum | +guard elevator | 66.20 | 0.51 | 8593 |
| Maximum | +vial holder & guard detector | 63.98 | 0.43 | 9520 |

TABLE 3A
$^{3}H$ analysis in benzene PPO (6gm/L) POPOP (0.2gm/L) (optimized regions)

| Amount of pulse index | Sample | $^{3}H$ efficiency | Background | $E^2/B$ |
|---|---|---|---|---|
| None | Sample only | 54.71 | 10.99 | 272 |
| Maximum | Sample only | 50.94 | 3.05 | 851 |
| Maximum | +vial holder | 49.38 | 1.37 | 1778 |
| Maximum | +guard holder | 38.44 | 0.86 | 1718 |
| Maximum | +vial holder & guard detector | 37.13 | 0.79 | 1745 |

TABLE 3B
$^{3}H$ analysis in water (large volume), (optimized regions) - 8ml sample, 12ml Pico-Fluor LLT

| Amount of pulse index | Sample | $^{3}H$ efficiency | Background | $E^2/B$ |
|---|---|---|---|---|
| None | Sample only | 22.85 | 6.12 | 5460 |
| Maximum | Sample only | 25.39 | 6.63 | 6223 |
| Maximum | Sample only | 25.63 | 3.87 | 10863 |
| Maximum | +guard detector | 23.77 | 2.29 | 25791 |

## CONCLUSION

Our $^{14}C$ and $^{3}H$ data clearly show that the plastic vial holder and/or guard detector technique increases the performance of the low-level liquid scintillation analyzer. For $^{14}C$ $E^2/B$ increased 2 to 2.5-fold using the plastic scintillator guard and/or holder. For the $^{3}H$ samples, performance increased by 1.5 to 2-fold using these plastic scintillators. Thus extremely good performance is obtained by using a plastic vial holder and/or guard detector in conjunction with afterpulse discrimination. Thus low-level liquid scintillation analysis can be used for detection of adulterants, $^{14}C$ age dating, low-level tritium in water, and monitoring of many other radionuclides in the environment, without requiring special, dedicated instruments. A general-purpose liquid scintillation analyzer with a low-level feature can analyze both low-level and routine laboratory samples for DPM determinations.

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