SELECTABLE DELAY BEFORE BURST - A NOVEL FEATURE TO ENHANCE LOW-LEVEL COUNTING PERFORMANCE

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ABSTRACT. Time-Resolved Liquid Scintillation Counting (TR-LSC™) has been successfully applied to environmental analysis, radiocarbon dating, nuclear power plant monitoring and food adulteration studies. Selectable delay-before-burst is a TR-LSC™ enhancement that improves sensitivity. User-selectable settings are available to adapt TR-LSC™ to new cocktails and higher-energy radionuclides. Significant increases in figure of merit (E^2/B) are achievable in certain applications. We present results correlating the increases in E^2/B with different delay-before-burst settings when counting ^{14}C and other environmental radionuclides in various cocktails. We discuss the significance of this increase in performance for various applications.

INTRODUCTION

Time-Resolved Liquid Scintillation Counting (TR-LSC™) is a patented technique used to discriminate unquenchable from quenchable background. Unquenchable background is caused by events that occur outside of the scintillation solution. These unquenchable events derive from cosmic radiation, external radioactive sources and natural radioactivity in the counting vial wall and the photomultiplier surface. The radiation interacts with the glass envelope of the photomultiplier tube (PMT), the vial wall and other material surrounding the vial to produce low-photon-yield Cerenkov events. Quenchable background, on the other hand, results from interactions of radiation with the LS solution. The light pulses produced from this type of interaction result in scintillation pulses similar to pulses induced by beta decay. Most of these quenchable events are from higher energy radiation because the particles must penetrate the lead shielding, the reflector and the glass wall of the vial before interacting with the LS cocktail. About 32% of the total background spectrum is due to quenchable events, whereas 68% is due to unquenchable background events. TR-LSC™ eliminates the unquenchable component of the background based on the differences in the characteristic light pulses from the quenchable and unquenchable components.

Programmable TR-LSC™ (selectable delay-before-burst counting) is used to enhance counting efficiency for radionuclides with β energies greater than ^3H, and when using newer generation cocktails with long decay constants. To understand why programmable TR-LSC™ is needed, we must characterize the differences between quenchable and unquenchable scintillation pulses.

DESCRIPTIVE BACKGROUND

Discriminating Between Quenchable and Unquenchable Background

TR-LSC™ is effective in eliminating the unquenchable component because the unquenchable component light pulses (Cerenkov photons) are different from those produced by the interaction of radiation with the scintillation solution. A true scintillation pulse (quenchable event) results from the transfer of kinetic energy from the incident radiation to the solvent and fluors of the scintillation solution, and is composed of a prompt and delayed component (Fig. 1A). Figure 1A does not represent the pulse shape of a single decay; rather, it represents the average pulse shape of a large number of decays over time (Roessler, Valenta & van Cauter 1989). Most of the light emitted is due to the prompt component (the lowest excited singlet state), and results in a flash of light

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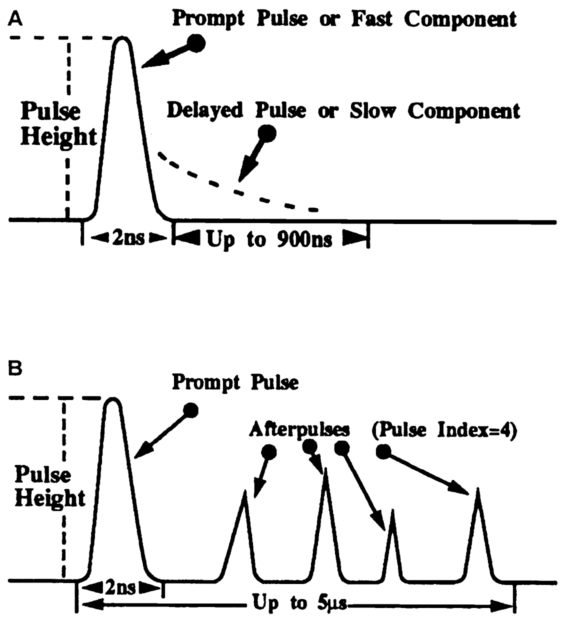

Fig. 1. A. Typical cocktail photoluminescence decay curve. B. Typical pulse pattern due to unquenchable background

(scintillation) of 2–8 nsec, which is proportional in intensity to the energy of the particle. The delayed component is caused by delayed fluorescence emission (as a result of the decay of two triplet excited molecules) in the scintillation cocktail, and persists for several hundred nanoseconds (Laustriat 1968). Dissolved O_2, which is present in samples prepared under normal laboratory conditions, reduces this delayed component.

In contrast, an unquenchable background pulse is composed of a prompt component followed by a delayed component consisting of a burst of afterpulses (single photoelectrons), which are fewer in number and spread out over time (Roessler, Valenta & van Cauter 1989). This burst of afterpulses can continue for up to 5 μsec (Fig. 1B). These trailing afterpulses have been reported by other investigators (Dressler & Spitzer 1966; Jerde, Peterson & Stein 1967; Roodbergen, Kroondijk & Verhud 1972). Usually, a far greater number of afterpulses for a given energy will result from an unquenched background event than from a true scintillation event. As a result, TR-LSC™ can be used to discriminate these background events. When a cosmic particle passes through the system, it can be discriminated against by counting the number of afterpulses produced (burst counting). In this technique, each coincidence event opens a burst-counting window, which counts the number of afterpulses occurring for about 5 μsec after the event. The total number of afterpulses is defined as the pulse index. The pulse index is used to create a three-dimensional spectral plot containing time-resolved information for each detected scintillation event (Fig. 2A). Figure 2A shows that a background sample gives an appreciable number of afterpulses at the low-energy end of the spectrum. In contrast, an unquenched ^3H sample generates few afterpulses and only at the high-energy end of the spectrum (Fig. 2B). An air-quenched ^3H sample (Fig. 2C) generates almost no afterpulses. By accepting only counts with few afterpulses, a reduced-background spectrum can be obtained.

TR-LSC™ may reduce counting efficiency for some radionuclides. In some cases, sample events will cause afterpulses, leading to rejection of a valid count event. This is especially true of deoxygenated samples and long-lived scintillators that have a significant delayed component. In ad-

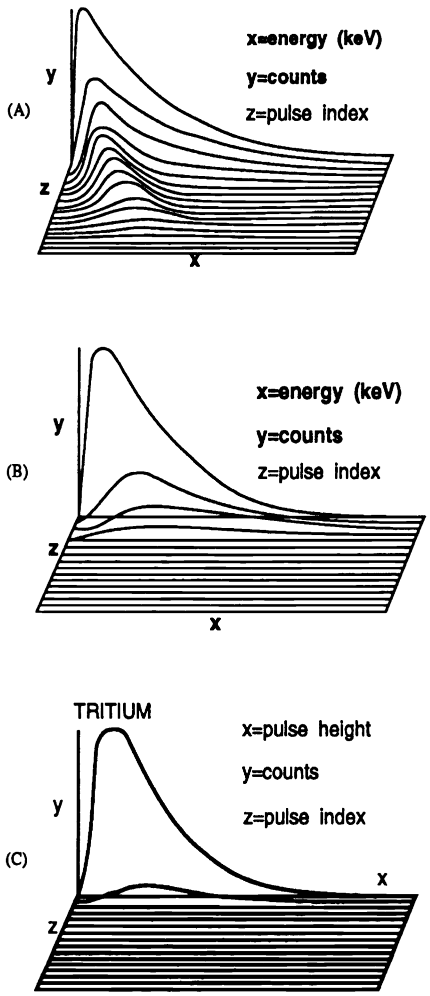

Fig. 2. Three-dimensional plot of pulse-height spectrum of background sample. A. The pulse index is the third dimension. B. Three-dimensional spectrum of an unquenched ^{3}H sample. C. Three-dimensional spectrum of an air quenched ^{3}H sample

dition, higher energy radionuclides (energies $\geq ^{14}C$) will produce more persistent delayed components, even if the sample is oxygenated. Programmable TR-LSC™ provides the ability to adjust the burst-counting window by increasing the delay before afterpulses are counted.

Programmable TR-LSC™ Performance

Cocktail chemistry and nuclide energy affect the persistence of the delayed component of a scintillation pulse. The newer-generation cocktails contain solvents that prolong the duration of scintillation pulses by enhancing the contribution of the delayed component. In addition, the delayed component becomes more prominent with increasing radionuclide energy. The programmable TR-LSC™ feature minimizes the counting efficiency loss and thus enhances sensitivity by allowing an increase in the delay before afterpulses are counted. Figure 3 graphically represents programmable TR-LSC™. This feature allows the user to adjust the delay from the system default setting of 75 nsec to a maximum of 800 nsec.

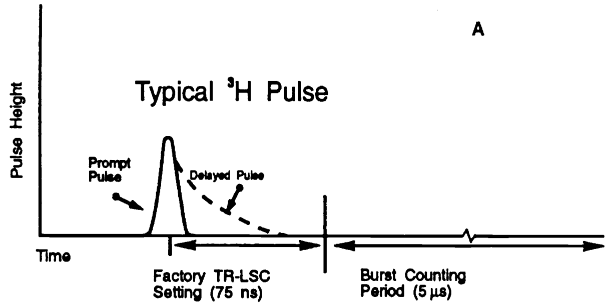

Fig. 3A. Conventional TR-LSC™

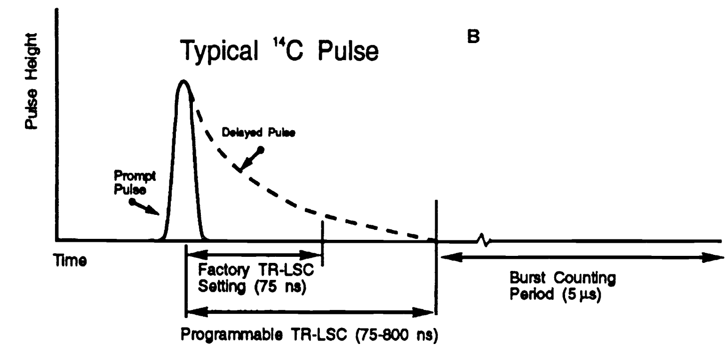

Fig. 3B. Programmable TR-LSC™

Methods

We performed a series of experiments with ^{3}H, ^{14}C, and $^{90}Sr/^{90}Y$ to assess the benefits of programmable TR-LSC™. The ^{14}C and $^{90}Sr/^{90}Y$ samples were prepared in both Insta-Gel® XF and Ultima Gold™ XR cocktails. The ^{3}H sample was prepared in Pico-Fluor™ LLT. All of the samples were prepared in 20-ml glass vials and contained 10 ml of water and 10 ml of the appropriate cocktail. Background samples were also prepared with 10 ml of ordinary distilled water and 10 ml of the appropriate cocktail. The ^{3}H sample contained 833 Bq (50,000 dpm), the ^{14}C samples contained 908 Bq (54,500 dpm), and the $^{90}Sr/^{90}Y$ samples contained 550 Bq (33,000 dpm). The overall error in the activity value is ± 2% in all cases. All samples were counted in a Tri-Carb® 2550TR/AB low-level α/β system using the low-level count mode. The programmable TR-LSC™ feature was used to vary the burst-counting window from the system default setting (75 nsec) to a maximum of 800 nsec in 100-nsec increments. All samples exhibiting activity were counted to a 0.5% 2 σ level of accuracy. Backgrounds were counted for 60 min. All samples were counted in wide regions from 0 keV to the energy maximum (E_{max}) of the isotope, as well as in an optimized region. We report data here for optimized regions because, in most low-level applications, region optimization is used to provide the maximum sensitivity.

Results and Discussion

The optimum settings of the delay-before-burst time window for each radionuclide in the appropriate cocktail was determined empirically by counting the samples at incremental delay-before-burst settings. The optimum delay setting results in the highest E^2/B.

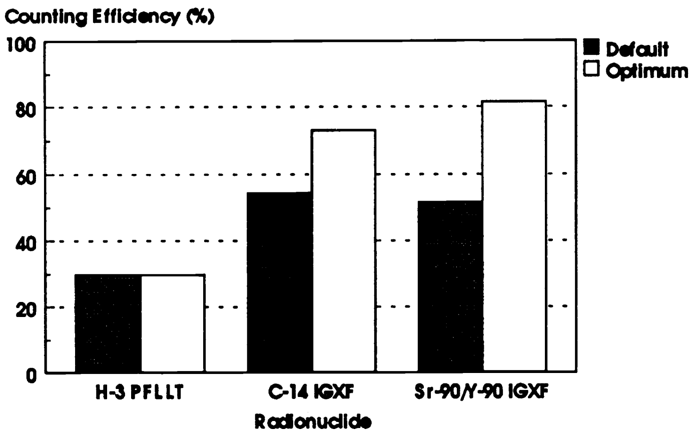

Fig. 4. Counting efficiencies at the default and optimum TR-LSC™ settings

Figure 4 shows the effect on counting efficiency for the various isotopes. The data presented show that a significant improvement in counting efficiency is achieved for both ^{14}C and $^{90}Sr/^{90}Y$. Counting efficiency for ^{14}C and $^{90}Sr/^{90}Y$ is improved because of the ability to compensate for the afterpulse effect of the delayed component of the scintillation pulse produced by these radionuclides. A small increase in the delay time before afterpulses are counted significantly increases counting efficiency.

^{3}H shows a small increase in efficiency at the maximum time delay setting (29.7%–31.7%). However, the optimum delay setting for ^{3}H, at the maximum E^2/B, is the system default setting. Thus, Figure 4 shows no improvement for ^{3}H counting efficiency. Although ^{3}H Pico-Fluor™ LLT data are presented, studies have shown that the counting efficiency for ^{3}H samples counted in Ultima Gold™ XR also show a small increase (22.8% *vs.* 25.8%) at the system maximum delay time. However, the optimum E^2/B for ^{3}H counted in both cocktails is at the system default setting (Fig. 5).

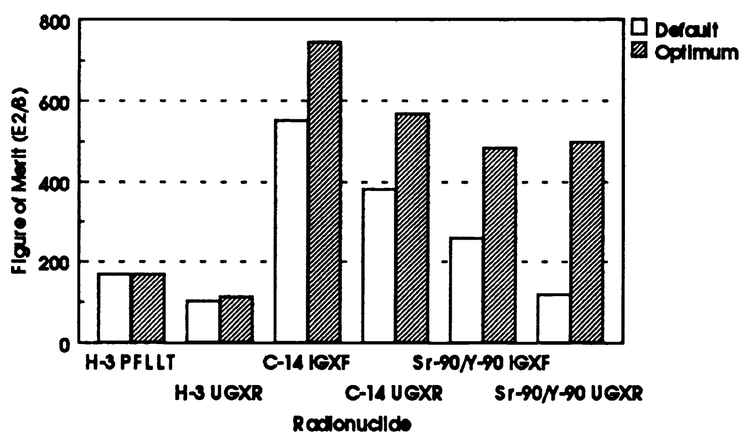

Fig. 5. Figure of merit at the default and optimum TR-LSC™ settings

Figure 5 shows a 135% increase in E^2/B for ^{14}C and an 187% increase for $^{90}Sr/^{90}Y$ in Insta-Gel® XF at the optimum burst delay time. A 148% and a 426% increase are seen respectively for both isotopes in Ultima Gold™ XR. The greater E^2/B increases in Ultima Gold™ XR cocktail are attributed to the fact that increasing the delay-before-burst counting minimizes the effect of tailing of the prompt scintillation pulse with this slower scintillating cocktail. The improvements in E^2/B for

^{14}C and $^{90}Sr/^{90}Y$ are due to the increase in counting efficiency realized at the optimum TR-LSC™ setting. There is no further background reduction at the optimum setting.

The benefit of an increase in E^2/B is obvious when the time required to achieve a desired minimum detectable activity (MDA) is considered. The lower limit of detection is calculated as

$$MDA = \frac{4.66\ B^{1/2}}{EVT \times 60} \quad (1)$$

where

MDA = minimal detectable activity in Bq ml^{-1}
B = total background counts accumulated
E = counting efficiency (decimal equivalent)
V = sample volume in ml
T = count time in minutes.

This formula assumes that the background and the sample are counted for the same amount of time. Figure 6 shows the time required to achieve an MDA of 1.85 Bq $liter^{-1}$ (50 pCi $liter^{-1}$) for ^3H, ^{14}C and $^{90}Sr/^{90}Y$. We chose 1.85 Bq $liter^{-1}$ (50 pCi $liter^{-1}$) as a limit because this is a guide value recommended in the U.S. Environmental Protection Agency (EPA) National Interim Primary Drinking Water Regulations. Many samples with gross β activity of <1.85 Bq $liter^{-1}$ (50 pCi $liter^{-1}$) do not require additional analysis because the sample is in compliance. Except for ^3H, Figure 6 shows that choosing the optimum delay time setting can result in a significant reduction in the count time required to achieve the same MDA.

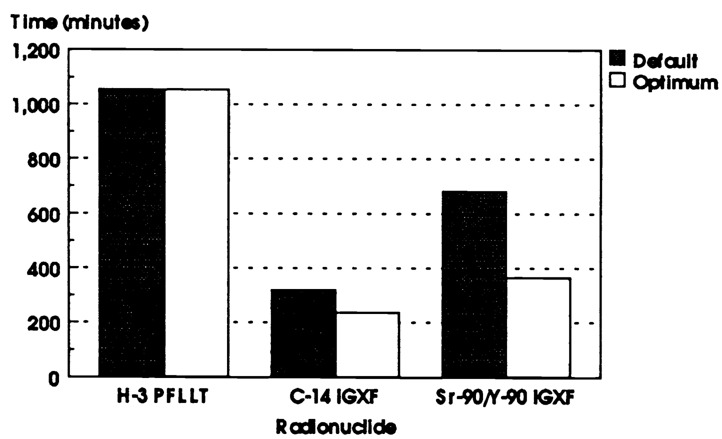

Fig. 6. Time required to achieve an MDA of 1.85 Bq $liter^{-1}$ (50 pCi $liter^{-1}$) at the default and optimum TR-LSC™ settings

Conclusions

The addition of digital time-resolved techniques to LSC has already resulted in technical innovations that advance the state-of-the-art in low-level counting for ^3H and ^{14}C analyses. Until now, to obtain optimum performance for ^{14}C and other higher energy radionuclides, such as $^{90}Sr/^{90}Y$, cocktail modification was required to reduce the delayed component of their scintillation pulses (Cook *et al.* 1989). Programmable TR-LSC™ provides: 1) user-selectable modification of the instrument rather than the cocktail. Improved performance (E^2/B) may now be obtained with a variety of cocktails, without modifying the cocktail; 2) improved TR-LSC™ performance for higher energy radionuclides. Conventional TR-LSC™ provides 2-to-3-fold increases in E^2/B for radionuclides such as ^{14}C, ^{35}S and ^{45}Ca. Programmable TR-LSC™ further improves performance 1.5–2 times. Because of this improvement, lower MDA values are achieved in a shorter time, which increases throughput. Programmable TR-LSC™ increases the field of applications for low-level β LSC.

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