Time resolved liquid scintillation counting in Tri-Carb and Quantulus GCT models from Revvity.

## Introduction

The invention of time-resolved liquid scintillation counting (TR-LSC $^{\text{TM}}$ ) technology led to a new dimension in sensitivity for Tri-Carb $^{\text{TM}}$  scintillation counters. Traditional liquid scintillation counting (LSC) uses coincidence counting to distinguish background events from true scintillation events. While this technology successfully eliminates much of the background counts due to electronic noise, it does not eliminate instrument background, arising from cosmic interactions with the glass face of the PMT or counting vial.

This cosmic component and noise can cause up to 70% of the remaining background counts. Revvity uses a combination of the traditional coincidence counting and a patented technology named time-resolved liquid scintillation counting (TR-LSC) to distinguish true decay events from background events. LSC sensitivity is a function of sample counting efficiency and background. Higher efficiency and lower backgrounds dramatically increase counting sensitivity. TR-LSC technology reduces background by as much as 97%, thereby, greatly increasing sensitivity for all LSC applications.

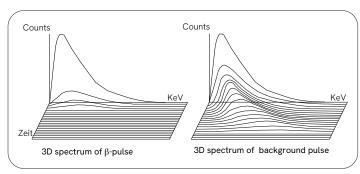
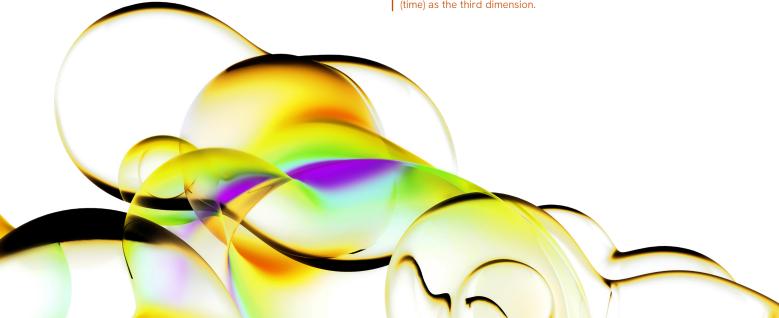


Figure 1: 3-D Display of the pulse height spectrum with pulse index (time) as the third dimension.



# Why is TR-LSC superior to conventional liquid scintillation counting?

The characteristics of  $\beta$ -scintillation pulses and background radiation pulses are substantially different. Both consist of a prompt or fast component, but a background pulse is followed by a series of low intensity afterpulses up to 5  $\mu s$  after the prompt pulse.

TR-LSC, adds a third dimension, called the pulse index, to liquid scintillation. The pulse index is a measure, over time, of the afterpulses associated with background. With this 3-D spectral analysis (see figure 1 and 2) TR-LSC, distinguishes true  $\beta$ -decay from background events, leading to increased sensitivity and superior counting performance.

Furthermore, TR-LSC is programmable to optimize counting any  $\beta$ -emitter in any cocktail. TR-LSC afterpulse discrimination is adjustable to accommodate the delayed component that often accompanies higher energy  $\beta$ -scintillation pulses to prevent this delayed component from being misclassified as background. The programmable feature of TR-LSC minimizes the misclassification of the b-activity as background, resulting in higher counting sensitivity for higher energy b-emitters such as  $^{90}\text{Sr}/^{90}\text{Y}$ . The QuantaSmart Software for Tri-Carb scintillation counters allows the adjustment of the delay before burst from 75ns (default value) to 800ns.

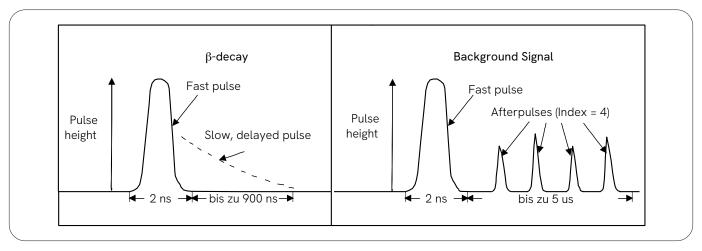


Figure 2: Typical pulse from  $\beta$ -particle and background.

Different sample types and counting conditions require more or less counting sensitivity. For low count rate samples, background becomes increasingly important. To accurately determine the net count rate of a low activity sample in a reasonable time, the background count rate must be small relative to the sample count rate. Revvity offers liquid scintillation detectors with four count modes, which use increasing levels of pulse index discrimination (TR-LSC), and provide faster and more accurate results for samples of virtually any activity.

# Increase of $E^2/B$ up to 10 times for better sensitivity

The ratio of the squared counting efficiency (E) and the background (B) signal is usually called the "Figure of Merit" and is a measure of the sensitivity of the liquid scintillation

counter. Figure 3 shows a comparison of the four possible count modi available with Tri-Carb scintillation counters for the isotopes  $^3\mathrm{H}$  and  $^{14}\mathrm{C}$ .

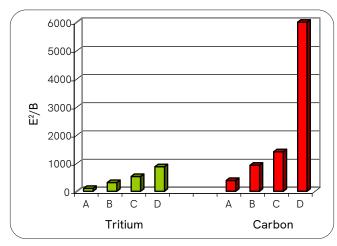


Figure 3: Relative  $E^2/B$ -values for TR-LSC count modes for  $^3H$  and  $^{14}C$  (as measured at the factory in Downer's Grove, IL, USA).

### Legend:

- A. Normal count mode (NCM) is standard on all Revvity Tri-Carb systems and reduces background by more than 20-30% versus conventional liquid scintillation counters (LSC). Recommended for samples above 500 CPM.
- **B.** High sensitivity count mode (HSCM) is for low activity samples and reduces background by more than 40% versus conventional LSC. Recommended for samples between 50 and 500 CPM.
- C. Ultra low level count mode (ULLCM) reduces background by more than 70% versus conventional LSC. Recommended for samples with less than 50 CPM.
- D. Super low level count mode (SLLCM) is standard on all Revvity 3180TR/SL models and employs a patented BGO (bismuth germanium oxide) background detector guard that surrounds the sample. It reduces background by more than 90% versus conventional LSC. This surround TR-LSC technology provides the highest sensitivity of any commercial LSC. Recommended for extremely low activity samples, such as in radiocarbon dating studies or environmental <sup>3</sup>H.

Table 1: <sup>3</sup>H Performance data gathered using 8 ml water and 12 ml Ultima Gold™ LLT in glass vials. <sup>3</sup>H Energy window was 0-18.6 KeV.

TR-LSC Mode	% <sup>3</sup> H Efficiency	Background (CPM)	E²/B
None	34.2	22.3	52
Normal	33.8	17.8	64
HSCM	33.9	12.8	90
ULLCM	29.4	6.2	139
SLLCM	26.7	2.8	255

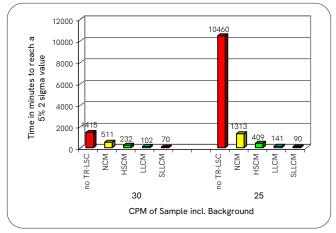


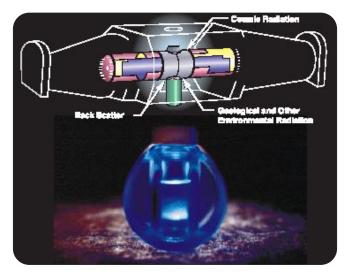
Figure 4: Effect of TR-LSC on the time required to achieve 5%  $2\sigma$  accuracy. Background sample = 12 ml Ultima Gold<sup>TM</sup> LLT, 8 ml "dead" water, in Glass Vials, <sup>3</sup>H window 0-18.6 KeV.

#### Surround TR-LSC in Quantulus™ GCT 6220

Revvity offers a dedicated low level counter, which combines TR-LSC technology with Bismuth Germanate (BGO) sample guards, creating Surround TR-LSC, to provide the ultimate in low activity sample detection. Surround TR-LSC delivers the lowest background of any multipurpose LSC. Not only is Surround TR-LSC sensitivity unsurpassed, but it also eliminates the bulk and complexity of anticoincidence guards, and massive lead shielding.

BGO sample guard technology allows you to use ordinary, disposable glass or plastic vials, while still achieving the highest level of sensitivity. The BGO guard closely surrounds the sample vial, rejecting background events before they interact with the sample. This eliminates the need to use expensive vials made of copper and Teflon®, or quartz, designed specifically for low level counting in other systems.

This also overcomes laborious washing and memory problems encountered with these special vials. With Surround TR-LSC you simply discard your vials when assay analysis is completed.



| Figure 5: BGO-Detector Guard in Quantulus GCT 6220.

Table 2: Achieve a target sensitivity for <sup>3</sup>H with either large or small volume samples with Surround TR-LSC. Data acquired and calculated at Ontario Power, Whitby, Ontario.

Sample (ml water)	Cocktail* (ml)	Region of interest (KeV)	Efficiency (%)	Background (CPM)	Count times (minutes)	Target sensitivity (Bq ³H/L)
3**	3	0.0-4.0	19.50	0.64	470	5.0
10**	10	0.0-4.0	19.89	0.87	60	5.0

#### | Table 3: 14C dating using the 3180TR/SL.

Sample/Vial type	Efficiency (%)	Background (CPM)	E <sup>2</sup> V <sup>2</sup> /B	Minimum age*** (years)
2g Benzene/7 ml glass	64.8	0.31	54181	48900
4g Benzene/7 ml glass	63.0	0.41	154888	49300

<sup>\*</sup> Ultima Gold LLT Cocktail.

## Radon analysis in drinking water

Liquid Scintillation is an approved EPA method (EPA Method 7500-Rn) for measuring radon in drinking water. The EPA has proposed concentration limits as low as 300 pCi/L. Revvity Tri-Carbs easily achieve this detection level of 0.66 Bq/L

or less in 60 minutes counting time. Tri-Carbs equipped with the alpha/beta discrimination feature can achieve a detection limit of 0.21 Bq/L in 60 minutes (see also Table 4 and 5).

| Table 4: <sup>222</sup>Rn results<sup>†</sup> for conventional LSC method in 10ml Cocktail with 10ml sample and 60 minutes counting time.

Cocktail	Optimized Region (KeV)	Efficiency* (%)	Background (CPM)	MDA (Bq/L)	MDA (pCi/L)
Mineral Oil Scintillator	200-600	320	3.47	0.60	16
Ultima Gold F	300-950	315	4.00	0.66	18
Opti-Fluor O	250-750	305	4.01	0.68	18
Insta-Fluor	250-750	300	3.72	0.67	18

### | Table 5: $^{222}$ Rn Results for $\alpha/\beta$ -LSC method in 10ml Cocktail with 10ml sample and 60 minutes counting time.

Cocktail	Optimized Region (KeV)	Efficiency* (%)	Background (CPM)	MDA (Bq/L)	MDA (pCi/L)
Ultima Gold F	300-950	243	0.200	0.21	6
Opti-Fluor O	250-750	240	0.867	0.42	11
Insta-Fluor	250-750	224	0.700	0.41	11
Mineral Oil Scintillator	200-600	219	1.020	0.49	13

<sup>\*</sup> Alpha-efficiency calculated as percentage of total radon and radon daughter emissions.

<sup>\*\*</sup> Revvity polyethylene vial (22 ml).

<sup>\*\*\*</sup> Based on minimal detectable amount (MDA) in DPM/g carbon which corresponds to A and is calculated using 2,71+4,65 ( $N_{\rm Bkg}$ ) $^{1/2}$  /T where  $N_{\rm Bkg}$  is the total number of background counts and T is the total count time (2000 minutes). This expression was given by Currie 1968.  $A_0 = 12,5$  dpm/g,  $t = 1/\lambda \ln(A_0/A_0)$ .

<sup>†</sup> All results reproduced with permission of Dr. Michael Cantaloub, Waste Management Federal Services, Richland, WA, USA.

### Alpha/Beta measurements

Many samples for environmental monitoring, nuclear power station monitoring, drinking water analysis, and nuclear waste clean-up, require analysis of total beta and total alpha radionuclides. However, in liquid scintillation counters, the pulse height spectra of alpha emitting radionuclides overlap the spectra of higher energy beta emitters, such as <sup>137</sup>Cs, <sup>89</sup>Sr and <sup>90</sup>Sr/<sup>90</sup>Y. Therefore, it is necessary to employ a form of pulse shape discrimination to differentiate alpha from beta emissions and compensate for this interference.

Revvity's time resolved Pulse Decay Analysis (TR-PDA), differentiates alpha from beta pulses according to their pulse decay characteristics. The pulse duration for alpha particles is typically longer than for betas. TR-PDA can run simultaneously with low level count modes, thereby achieving extremely low misclassification of beta and alpha particles, without compromising sensitivity.

TR-PDA provides automatic or manual adjustment of the pulse decay discriminator (PDD). The automatic alpha/beta standard mode scans the range of possible PDDs without operator intervention, for both alpha and beta pulses, and determines the optimum PDD for minimum spill over. In the manual alpha/beta mode, the range and number of PDD values are set by the operator.

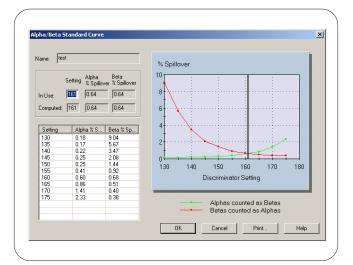


Figure 6: An example of  $\alpha/\beta$ -misclassification curve from the Tri-Carb  $\alpha/\beta$ -standards library.

## Sample oxidizer

Revvity's sample oxidizer makes it easy to prepare many samples containing <sup>3</sup>H and <sup>14</sup>C. Organic matter in vegetation, animal tissue, solid waste, and even soil is easily and rapidly oxidized into <sup>3</sup>H<sub>2</sub>O and <sup>14</sup>CO<sub>2</sub>, and completely prepared for counting. After combustion <sup>3</sup>H and <sup>14</sup>C are physically separated, therefore, dual label analysis is simplified considerably. Revvity sells a complete lone of reagents to ensure superior performance and consistency.

#### **TR-PDA Benefits**

- Uses time resolution technology to analyse pulse decay characteristics and discriminate alpha/beta decay events.
- Automatic optimization of  $\alpha/\beta$ -separation and counting, minimizing misclassification in a variety of counting samples.
- Settings can be optimized and configured independently for each protocol and stored for application to any other counting protocol.

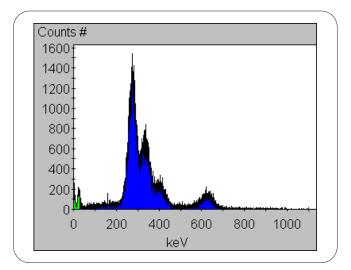


Figure 7: Radium, Radon and Polonium daughter nuclides measured in Ultima Gold LLT.

## LSC Cocktails1)

## Ultima Gold LLT for Low Level applications

Ultima Gold LLT is suitable for the direct determination of low levels of <sup>3</sup>H in a wide range of water samples. Direct addition removes the need for distillation. Ultima Gold LLT accepts up to 54% distilled water, deionized water, tap water, rain water, river water and has a large capacity for sea water, with 3H counting efficiencies of approximately 26% and low backgrounds. When used with Revvity's TR-LSC technology minimum detectable activities of <1.1 Bq/L are attainable (see also Table 6).

## Ultima Gold AB for $\alpha/\beta$ -samples

Ultima Gold AB was specifically designed for use in alpha/beta separation. The use of a slow pulse decay solvent with optimized detergent type and concentration combine to produce a cocktail with all the characteristics necessary for effective alpha/beta separation. The outstanding sample holding capacity of Ultima Gold AB for aqueous and acidic sample types makes it the cocktail of choice for most alpha/beta LSC applications (results see also Table 7).

Table 6: Performance of Ultima Gold LLT for low level <sup>3</sup>H using a Tri-Carb 3180TR/SL in a counting window from 0.4-4.5 KeV. Results generated at the Scottish Universities Research and Reactor Center, East Kilbrigde, Scotland.

Water to cocktail ratio	Efficiency ³H (%)	Background (CPM)	E²/B	E²V²/B	MDA (Bq/L)
8ml : 12ml	24,6	1,15	526	33680	1,22
10ml : 10ml	21,2	1,11	405	40490	1,11
11ml : 9ml	18,1	0,95	345	41730	1,06

Table 7: Performance of Ultima Gold AB with samples at 20°C using a low level Tri-Carb equipped with alpha/beta discrimination (0-2000 keV region of interest).

	Water	1.0 M HCl	2.0 M HCl	1.0 M HNO <sub>3</sub>	2.0 M HNO <sub>3</sub>
Sample uptake capacity	10.0 ml	5.0 ml	2.25 ml	3.25 ml	2.25 ml
Missclassification	1.8%	1.7%	1.0%	3.8%	4.2%

Every Revvity Tri-Carb delivers verified, reliable results. That's because every Tri-Carb employs patented TR-LSC background reduction; double light seals for maximum PMT stability, low energy <sup>133</sup>Ba external standardization for the best quench monitoring; and safe, gravity assisted sample downloading to prevent vial jamming and breakage.

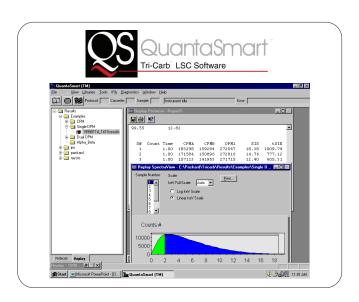
#### Instruments and electronics

- Linear MCA.
- Patented, programmable TR-LSC background reduction.
- Powerful integrated computer system control and data management.
- Optimized and matched photomultipliers.
- Luminescence detection.



#### QuantaSmart software

- Windows 10 operating system.
- SpectraBased counting of isotopes selected from radionuclide libraries.
- Validated instrument performance assessment (IPA) for compliance with the EPA's GLP recommendations and the FDA's GMP regulations.
- Automated calibration and normalization with sealed standards traceable to NIST (National Institute of Standards and Technology).
- 21 CFR part 11 compatibility.



## Analysis Software SpectraWorks<sup>2</sup>

- Real-time visualization and plotting of sample spectrum.
- Helps fine-tune regions for single, dual and triple label assays.
- Display and analyse unlimited number of spectra.
- Calculate detection limits.
- Confirms presence of luminescence.
- Provides evidence of quenching problems.

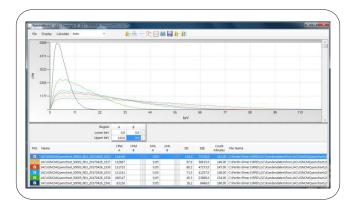


Table 8: Possible Tri-Carb/Quantulus models and available measurement modes, S = standard, O = optional, - = not available.

Model	NCM	HSCM	ULLCM	SLLCM	α/β
4810TR	S	-	-	-	-
4910TR	S	0	-	-	0
5110TR	S	-	S	-	S
6220	S	_	-	S	S

## Literature:

- Revvity (Germany) GmbH, 63110 Rodgau-Jügesheim, LSC Application Note, Cocktails for Liquid Scintillation Counting, October 2004.
- R. H. W. Edler; An Introduction to the Scintillation Technology for the Measurement of Radionuclides, 1<sup>st</sup> Edition, Bremen 2020, ISBN 978-3-00-020422-7.



