CHAPTER 16

Solid Scintillation Counting: A New Technique—Theory and Applications

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ABSTRACT

Solid scintillation counting is a new technique for measuring radioactive samples that previously had to be counted in liquid scintillators. The solid scintillator, XtalScint™, produces a light output brighter than standard organic scintillators. It is nonhazardous, nonodorous, and insusceptible to impurity (chemical) quench. It can be attached to either porous or nonporous surfaces for solution and filtration applications. Under optimum conditions, tritium efficiency approaches 60%. This report will discuss the theory and applications of XtalScint to solid scintillation counting techniques.

INTRODUCTION

Recent developments in liquid scintillation counting have focused on improving liquid scintillation solution safety by decreasing the volatility of the solvent (Kellogg,[1] Kalbhen and Tarkkanen,[2] Reed,[3] and Lin and Mei[4]). A solid, fine, scintillator powder that replaces the solvent would be the ultimate in low volatility. The powder, supported on either a porous or nonporous carrier, could be placed in a vial and counted in a liquid scintillation counter. The scintillation performance of the solid scintillation system would be equivalent to or superior to counting with liquid scintillation solutions. Also, in contrast to liquid scintillation solutions, the counting vials could be reused; the labeled samples could be recovered from the solid scintillators. Solids also have health and disposal advantages that make this new technique potentially revolutionary to the industry.

This report will discuss, in depth, the theory and applications of such a solid scintillation counting system using Beckman's Ready Cap and Ready Filter.

THEORY

There are five important properties of a scintillator. First, it must efficiently convert the energy of the radiation decay into measurable light. Second, it must be chemically inert to the conditions of measurement. Third, it must have low noise, or background. Fourth, it must be able to interact with low energy betas and augers. Finally, it should present minimal hazards to the user. XtalScint by Beckman meets all these criteria.

Conversion Efficiency

Figure 1 demonstrates that the light output for XtalScint is much greater than even an unquenched liquid scintillation calibration standard. Since these emission spectra were generated by the same isotope, this implies that XtalScint has a much greater light output per KeV of excitation energy than the best liquid scintillators. The wavelength of the emission maxima for XtalScint is 395nm, ideal for maximum sensitivity of current photomultiplier technology. The decay time of emission is 80 to 120 nsec. While this is slower than most traditional liquid scintillators, it is well below the minimum microsecond resolving power of current liquid scintillation counters and therefore very acceptable.

Chemical Properties

XtalScint is a solid with a melting point above 1000°C. It is chemically inert to aqueous buffers, aqueous bases, and organic solvents. It partially dissolves in concentrated acid, however, it can withstand moderate exposure to 1 *M* hydrochloric acid and 0.1 *M* sulfuric acid. Since XtalScint is completely impervious to organic reagents it is immune to impurity (chemical) quenching interferences. See Table 1 for data. It is also immune to chemiluminescence caused by common chemical reagents, such as base and peroxide.

Low Noise (Background)

XtalScint as used in Ready Cap and Ready Filter applications has essentially the background of an empty vial. Background generated from cosmic particle interaction with 10 mL of liquid scintillator is greatly reduced with Ready Cap or Ready Filter (see Table 2) because of the small scintillator target. Gillespie[5] has reported an 8- to 10-fold improvement in the signal to noise ratio for measurements of ^{32}P and ^{125}I with Ready Cap compared to liquid scintillator cocktail.

Detection Capability

While plastic scintillators and crystal scintillators are not new solid scintillators, they have found very little acceptance for measuring biological samples, most of which are labeled with tritium and ^{14}C. Although effective as scintilla-

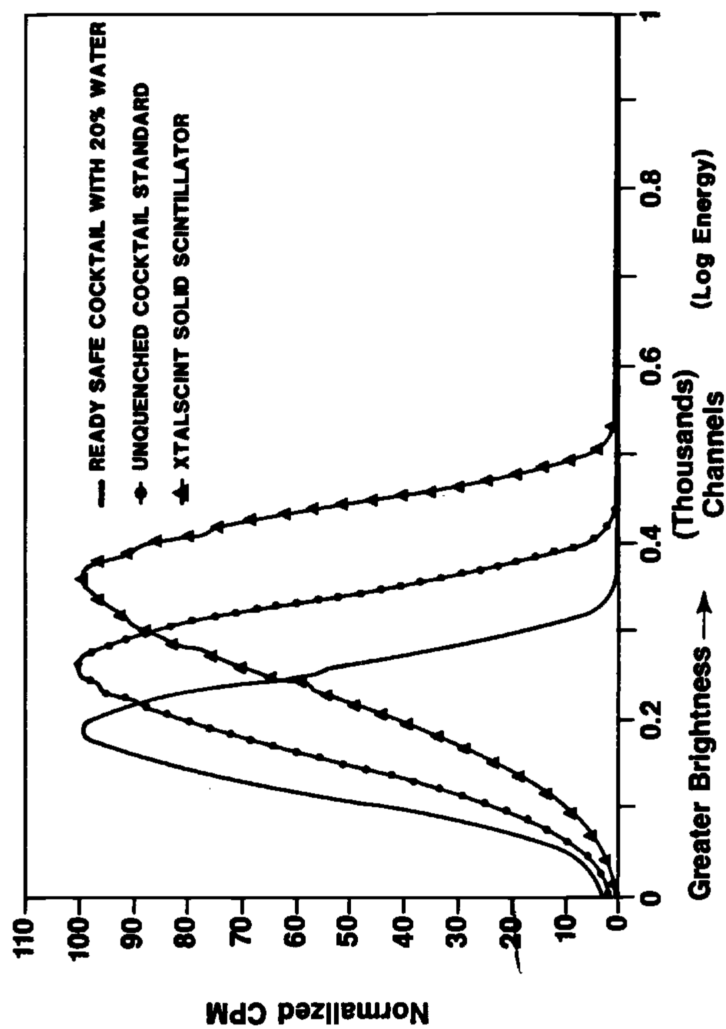

Figure 1. Light output (Tritium Spectra) with various scintillator forms.

Table 1. Effect of Chemical Quenching on Tritium Efficiency, at 20 nsec, of inulin in Liquid and Solid Scintillators

Quench Agent	Volume	Scintillator Liquid	Xtalscint
None	—	45.5%[a]	39% (DRY)[b]
Isopropanol[c]	100μL	45.0%	28.5% (WET)
Nitromethane[d]	100μL	2.9%	28.5% (WET)

[a]Tritiated inulin in water added to 5 mL of ready protein$^+$.
[b]Tritiated inulin in water added to ready cap, dried and counted.
[c]Isopropanol, 100μL, added to both cocktail and ready cap preparations. Samples were counted without removing isopropanol.
[d]Nitromethane, 100μL, added to both cocktail and ready cap preparations. Samples were counted without removing nitromethane.

tors, their poor contact with the labeled substrate has meant poor detection capability.

Two important developments make the XtalScint scintillator system effective. First, XtalScint is a fine (3 to 8 micron) powder providing enormous surface area for the intimate interaction between the labeled substrate and the scintillator. Second, Seltzer and Berger[6,7] and Unak[8] have calculated that by replacing the liquid media between scintillator particles with air, one increases the distance an electron can travel by a factor of 1000. Thus, removing water from an aqueous solution of ^3H 5-fluorouracil on ready cap resulted increased the tritium count rate from 13,700 to 75,900 cpm. This is a fivefold increase.

The performance of XtalScint may also be enhanced by altering the coincidence gate on the LS instrument. By increasing the coincidence gate time one can improve the tritium count rate by 25 to 40%. Enhancement of more energetic isotopes is not as significant (see Table 3).

Table 2. Background for XTALSCINT and Ready Safe

Sample	^3H 0–400	Window $^{14}C/^3H$ 401–670	$>^{14}C$ 671–1000
Empty Plastic Vial	14.43	14.17	0.33
10 mL Ready Safe in Plastic Vial	26.10	13.90	17.77
XTALSCINT, 35 mg, in Plastic Vial	14.55	14.06	0.53

Table 3. Radioisotope Efficiencies of XTALSCINT vs Ready Protein$^+$

Radioisotope	Substrate	Solvent	XtalScint 20 nsec GATE	XtalScint XTAL GATE	Ready PROTEIN
^3H	Palmitic Acid	2-Propanol	41.5%	57%	44%
^{14}C	Glutamic Acid	Water	93.4%	97.7%	94%
^{125}I	Triodothyroxane	2-Propanol	75%	78%	78%
^{32}P	Phosphoric Acid	Water	94%	98.5%	100%

Health and Safety Aspects

XtalScint has certain inherent laboratory safety advantages over liquid scintillators. XtalScint has no vapor, protecting the user from exposure to bad odors or toxic vapor inhalation. XtalScint is not absorbed through the skin, saving the user from possible internal organ damage. XtalScint does not need to be used in a hood, providing additional hood space previously occupied by liquid scintillators. Spilled XtalScint, should it come free from its carrier, can be cleaned up with a broom and dust pan rather than spill pillows and absorbent. XtalScint will not burn, in contrast to most liquid scintillators which are classified as flammable or combustible. XtalScint with carrier is smaller than liquid scintillators, making storage more efficient. XtalScint with carrier may be stored in any quantity; storage of hazardous liquid scintillators is proscribed by regulations.

Disposal Aspects

Solid scintillators such as XtalScint are much easier to dispose of than are liquid scintillators. Liquid scintillator waste must be packaged with an absorbent adequate to hold the spilled liquid in the event the scintillation vials or other waste containers should leak or break. As a result, a drum of liquid scintillator waste contains about 2/3 absorbent and only 1/3 actual vials. XtalScint, in contrast, is classified as dry waste and requires no supplemental absorbent. Because of its compactness (small size) and dry form, one drum of XtalScint samples on its carrier is equal to 30 to 300 drums of the same number of liquid scintillator samples in vials. This represents tremendous cost savings as well as stress relief for disposal sites and the environment in general.

SAMPLE PREPARATION AND COUNTING PROCEDURE

Ready Cap

Ready Cap is the cap of a small plastic vial coated with a layer of XtalScint[9] (see Figure 2). It is designed for counting nonvolatile, radiolabeled substrate in a volatile solvent. The liquid sample, less than 200 μL, is pipetted onto the scintillator surface in the Ready Cap. The volatile liquid is evaporated by numerous methods (evaporation in hood, heat lamp, hot air blow dryer, microwave oven, or vacuum centrifuge), leaving the labeled, nonvolatile substrate deposited on the surface of the scintillator. The Ready Cap is placed in an LS vial and counted on an LS counter. The Ready Cap can subsequently be removed from the vial and the vial reused. In many cases the radiolabeled substrate may be extracted and recovered for further analysis; this is not possible with liquid scintillator solutions.

The position of the cap in the vial makes very little difference to the measured count rate (see Table 4).

The labeled substrate may be any nonvolatile material, such as proteins,

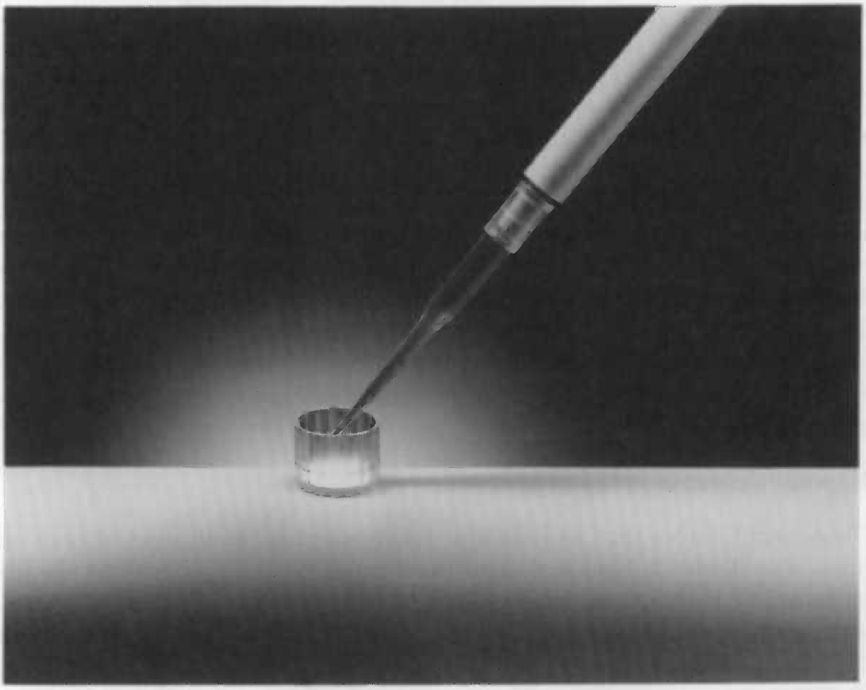

Figure 2. READY CAP: A small plastic container coated on the bottom with XtalScint.

peptides, amino acids, oligionucletides, steroids, etc., dissolved in a volatile liquid.

There are three limitations on the use of Ready Cap. First, the volume of the sample is limited to 200 μL. Second, because of the small target area of scintillator, it is difficult to generate a spectrum from an external gamma source for quench determination. Finally, noncorrectable quench is introduced by codeposition of nonradioactive, nonvolatile substances such as salts

Table 4. Ready Cap Position in Vial: Effect on Efficiency[a]

Ready Cap Position		Count Rate (CPM)	Relative to Bottom Down Configuration
Bottom Down	⊔	239,000	100%
Bottom Up	⊓	239,000	100%
Side Up Bottom Parallel to PMT Face	○	245,500	102.7%
Side Up Bottom Perpendicular to PMT Face	]	236,300	98.9%

[a]Single ready cap labeled with ^{3}H-5-Fluorouracil counted on four possible positions resting on bottom of plastic 20 mL Poly Q Vial.

Table 5A. Effect of Buffers on Ready Cap ^3H Counting Efficiency; Values Reported are Counting Efficiencies (%) at 20 nsec

Liquid Carrier for ^3H-Palmitic Acid	Volume of ^3H-Palmitic Acid Added			
	25 μL	50 μL	100 μL	200 μL
D.I. Water	41.8	39.6	35.8	33.0
0.1 *M* NaCl	42.7	40.3	37.1	33.2
1.0 *M* NaCl	41.7	38.0	32.8	23.3

(greater than 1 *M*) or high boiling solvents like glycerol, which absorb the beta decay before it reaches the scintillator (see Table 5a and 5b).

Ready Filter

Ready Filters are glass fiber filters coated on one side with XtalScint and designed to be used in a manner similar to conventional glass fiber filters. Ready Filter comes in two formats: filter mats for automated cell harvesters, and 25 mm filter circles for manual filtration experiments. Particulate suspensions are filtered onto the XtalScint side of the filter. To inactivate nonspecific binding, it may be occasionally necessary to presoak the filter in unlabeled substrate.

The flow rate of water through the Ready Filter is about 70% the flow rate through S&S #32 glass fiber filter. The retention of BSA precipitate for the two filters is greater than 99%.

Counting efficiency comparisons were made between Ready Filter and S & S #32 glass fiber filter counted in Ready Organic. The samples used contained 250 μg of BSA or DNA, labeled with different isotopes. The results of this comparison, reported in Table 6, show that the efficiencies of the two filters are similar to each other for ^3H, ^{14}C, ^{125}I, and ^{32}P labeled precipitates when counted on conventional liquid scintillation counters. Using a counter with an optimized coincidence gate yields superior tritium efficiency for the Ready Filter. Other isotopes are not as dramatically affected. Both filter types show similar patterns of efficiency dependence on substrate identity (Table 7) and similar weight of precipitate deposited (Table 8). Efficiency for Ready Filter was not affected by the vacuum caused increased flow rate through the filter (Table 9).

After drying Ready Filter, it may be counted in any vial, but the orientation of the filter in the vial makes a difference in counting efficiency (Table 10).

Table 5B. Effect of Buffers on Ready Cap Counting Efficiency; Values Reported AE Counting Efficiencies (%) at 20 nsec

Liquid Carrier for Amino Acid Mixture	Volume of ^{14}C-L-Amino Acid Mixture Added			
	50 μL	100 μL	10 μL	200 μL
D.I. Water	83.0	78.2	78.1	77.4
0.1 *M* NaCl	83.2	78.8	78.2	77.4
1.0 *M*	80.5	76.2	72.4	67.0

Table 6. Counting Efficiency of Common Isotopes: Ready Filter vs Standard Filter in Cocktail

Isotope	Substrate	Efficiency		
		Glass Filter[a]	Ready Filter[b]	Ready Filter[c]
^{3}H	BSA	32.6	30.1	36.6
^{14}C	BSA	96.9	87.3	92.0
^{125}I	BSA	57.8	62.2	70.8
^{32}P	LYSATE	101.6	87.35	95.5

[a]S & S #32 glass fiber filter counted in Ready Organic.
[b]Ready filter counted with 20 nsec gate.
[c]Ready filter counted with Xtal gate.

Table 7. Counting Efficiency as a Function of Substrate: Ready Filter vs Standard Filter in Cocktail

Substrate	Efficiency		
	Glass Filter[a]	Ready Filter[b]	Ready Filter[c]
^{3}H BSA	32.2	30.1	36.6
^{3}H Lysozyme	27.4	24.9	30.7
^{3}H DNA	29.2	30.2	36.3

[a]S & S #32 glass fiber filter counted in Ready Organic.
[b]Ready filter counted with 20 nsec gate.
[c]Ready filter counted with Xtal gate.

Table 8. Efficiency vs Weight of Precipitate: Ready Filter vs Standard Filter in Cocktail

Weight of ^{3}H BSA (μg)	Efficiency	
	Glass Filter[a]	Ready Filter[b]
5	35.1	36.5
50	43.1	41.0
100	37.4	37.2
250	35.8	35.5
500	32.0	32.0
750	31.5	29.6
1000	31.0	30.5

[a]S & S #32 glass fiber filter counted in Ready Organic.
[b]Ready filter counted with 20 nsec gate.

Table 9. Efficiency vs Flow Rate: Ready Filter vs. Standard Filter in Cocktail

Vacuum Measured in Inches of Hg Drop	Efficiency of ^{3}H BSA	
	Glass Filter[a]	Ready Filter[b]
6.8	30.6	33.1
10.0	29.3	30.0
15.0	30.3	30.4
20.0	32.9	32.9
25.0	32.2	32.2

[a]S & S #32 glass fiber filter counted in Ready Organic.
[b]Ready filter counted with 20 nsec gate.

Table 10. Ready Filter Position in Vial Effect on Efficiency[a]

Position of XTAL in Vial	Vial	Count Rate (CPM)	Relative to Maxi Up Configuration
Up	Maxi	52,500	100%
Down	Maxi	21,000	40%
In	Maxi	41,500	79%
Out	Maxi	39,000	75%
In	Mini	44,500	84%
Out	Mini	41,500	79%
In	Bio	38,000	72%
Out	Bio	39,500	75%

[a]Single ready filter with ^3H BSA precipitate counted in different orientations in three different vial types.

The best position for counting Ready Filter is flat on the bottom of the vial with the XtalScint side up. As with Ready Cap, the analyte can be recovered by extraction from the XtalScint and used for further analysis.

Ready Filter should not be used for counting nonvolatile substrate in true solution. The glass fiber support for Ready Filter is capable of strong, nonspecific binding. Upon drying, the nonvolatile substrate can be bound to the nonscintillating support and give reduced scintillations. An example of this is reported in Table 11. Precipitates on the other hand, are captured in the solid scintillator layer and give the high counting efficiencies reported. External quench monitors are not recommended with Ready Filter due to the time required to obtain the Compton spectra from such a small target.

CONCLUSIONS

XtalScint is an effective solid scintillator ideally suited for measuring ionizing radiation from radioactive decay. It may be measured on conventional liquid scintillation instrumentation, performing similar to conventional liquid

Table 11. Risks of Counting Labeled Substrate in Solution on Porous XtalScint Base (Ready Filter)

Solution	Efficiency[a]: Ready Filter	Efficiency[a]: Ready Cap
^3H Uracil	38.8%	39.1%
^3H Histamine	6.1%	39.5%

Scrap XtalScint from Ready Filter surface
Extract Filter Base and XtalScint with Ready Protein$^+$

	% Total DPM Extracted: Uracil	% Total DPM Extracted: Histamine
XtalScint from Ready Filter	52.4	7.4
Filter Base from Ready Filter	47.6	92.6

[a]Efficiency measured at 20 nsec gate.

scintillators, or measured on instruments with optimized coincidence circuitry (such as the Beckman LS 6000 series), performing better than conventional liquid scintillators. Its low background and high signal make it especially attractive for those experiments requiring high sensitivity.

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