CHAPTER 18

Some Factors Affecting Alpha Particle Detection In Liquid Scintillation Spectrometry

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ABSTRACT

Pulse shape analyzers have been introduced in commercial liquid scintillation counters, such as the Wallac 1220 QuantulusTM and 1219 SM, enabling separation of alpha particle spectra from other types of radiation in a single measurement and making ultra sensitive alpha counting possible. Background count rates as low as one count per day for ^{214}Po in a teflon vial are achievable while still retaining nearly 100% counting efficiency.

The number of photons per decay event however, is relatively low in liquid scintillation counting, leading to energy resolution of the order of 10%. Energy resolution has been found to improve with low water to cocktail ratios and opaque vial types.

Easy and fast sample preparation together with the ultra low background make liquid scintillation spectrometry a competitive method in environmental counting of alpha particles.

INTRODUCTION

Liquid scintillation counting (LSC) is a well established method for beta particle counting. Its use in alpha particle counting has suffered from large background count rates in energy range due to the cosmic ray and environmental radiation caused events in the cocktail. Background reduction for alpha counting requires identification of the origin of the background. It has long been known that the relative amounts of the prompt and delayed components of fluorescent decay depend on the specific ionization, hence the type of particle causing it. Typically, the heavily ionizing particles, such as neutrons and alpha particles, produce more delayed component than beta particles, thus longer pulses. This phenomenon is the basis for the pulse shape analysis (PSA) as particle identifier, and it makes possible rejection of the beta-like background component in alpha counting.

The method has been applied in nuclear physics for neutron counting in the presence of gamma radiation since the '50s. In alpha counting it has also been applied with good success and is well reviewed.[1,2]

Experimental liquid scintillation counters, developed in the '70s, used zero crossing techniques for pulse shape discrimination.[3,4] These devices contained silicon oil as a light guide, improving the coupling from the sample to a single photomultiplier tube, thus giving an excellent energy resolution.

Wallac 1220 Quantulus, an ultra low background liquid scintillation spectrometer, has contained a pulse shape analyzer since 1987. Being a general purpose alpha-beta counter with an automatic sample changing mechanism, the optical coupling described above was not applied, but a standard detector configuration with two PMTs was maintained.

The Wallac pulse shape analyzer uses a concept quite similar to the one adopted by, e.g., Brooks.[5] It initiates the integration of the pulse tail after 50 nsec from the leading edge and compares it with the integral of the total pulse. Surface mounted electronic components manufacture the high speed analog circuitry that produces the pulse shape related signal without amplitude dependence. By setting the PSA level through software commands it is possible to route alpha-like events into one half of the MCA and beta-like events (due to beta particles, Compton recoil electrons, Cerenkov radiation, and X-rays) into the other half in a single measurement. Maximum resolution of 1024 channels is available with a logarithmic energy scale.

The Quantulus is equipped with a guard counter, set in anticoincidence with the sample detector PMTs, to minimize the background component caused by environmental radiation, cosmic flux, and gamma flux from the bedrock and building materials. Further, a user programmable pulse amplitude comparator is available for reduction of cross talk events.[6]

Pulse shape separation characteristics depend on cocktail. Adding naphthalene to conventional toluene or xylene based cocktails enhances their pulse shape separation. Modern Hisafe cocktails by Wallac contain naphthalene derivatives as solvent and offer excellent pulse shape characteristics without any further naphthalene addition.

PULSE SHAPE ANALYSIS

Since the pulse shape characteristics vary between different cocktails, the pulse shape analyzer has to be set individually for each one to achieve the optimum separation of alpha and beta particle spectra. This can be done by preparing a pure alpha and beta sample whose amplitude spectra overlap, e.g., ^{36}Cl and ^{241}Am. The PSA level is then scanned for acceptable rejection of the radiation not of interest. With conventional cocktails one may allow say 5% loss of alpha particle counting efficiency to remove beta-like background, if alpha particle spectrum is only needed. If both particle types are to be measured, new cocktails containing naphthalene derivatives will give good separation (Table 1).

Table 1. Beta Residual, the Percentage of Betas Remaining Among Alpha-like Events. Normalized to 100% when PSA is off. The Residual is Given for ^{36}Cl Beta Particles in ^{241}Am Alpha Window for Corresponding Alpha Counting Efficiencies. The Sample is Contained in 1 mL Water with 10 mL Optiphase Hisafe 3 in Equilibrium with Air.

PSA	Alpha Eff %	Beta Residual %
0 = off	100	100
25	100	100
50	100	84.7
75	100	36.1
100	99.3	2.75
120	98.9	0.098
130	95.9	0.025
140	88.7	0.02
150	73.4	<0.02
160	54.4	

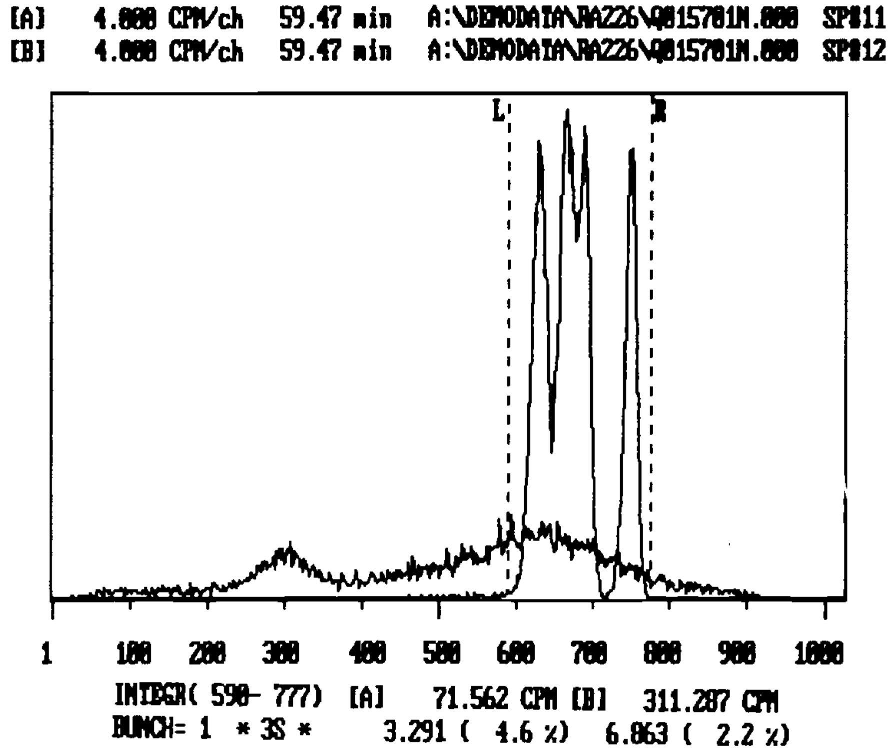
ENERGY RESOLUTION IN LIQUID SCINTILLATION SPECTROMETRY

Energy resolution is proportional to the number of photons created in the decay event.[7] With LSC one may not expect to achieve the energy resolution of solid state detectors, because number of liquid scintillator charge carriers is considerably greater than that of photons detected in a typical liquid scintillation event. Therefore, considerable improvement in energy resolution will only be achieved with a profound change in the light production properties of the cocktail.

Current cocktails have about 10% resolution in opaque vials at 5 MeV alpha energy (Figure 1). Corresponding resolution in standard glass vials is poorer, 20% (see item 2).

There are ways to improve the energy resolution:

1. High light output cocktails give better results.
2. Opaque vials transmit light with fewer losses than standard glass vials where photons can get trapped through total reflections in the vial wall. Teflon and etched glass have the best resolutions so narrower windows can be used to obtain smaller backgrounds with the same counting efficiency. Alignment of the liquid meniscus with the center axis of the PMT gives the best sensitivity and optimal resolution.
3. The lower the quench level, the better the photon emission; this leads to improved energy resolution. In practice this means having the smallest amount of water, contradicting the need to have as good a lower limit of detection as possible. In extractive methods, using two phase samples, radioactivity from water is transferred into the cocktail phase thereby eliminating the aqueous quench.
4. Nitrogen bubbling improves the performance further, due to the removal of quenching by dissolved oxygen.



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[A]   4.000 CPM/ch   59.47 min   A:\DEMODATA\RA226\Q015701N.000   SP#11
[B]   4.000 CPM/ch   59.47 min   A:\DEMODATA\RA226\Q015701N.000   SP#12
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INTEGR( 590- 777)  [A]    71.562 CPM [B]    311.287 CPM
BUNCH= 1  * 3S *       3.291 (  4.6 %)   6.863 (  2.2 %)
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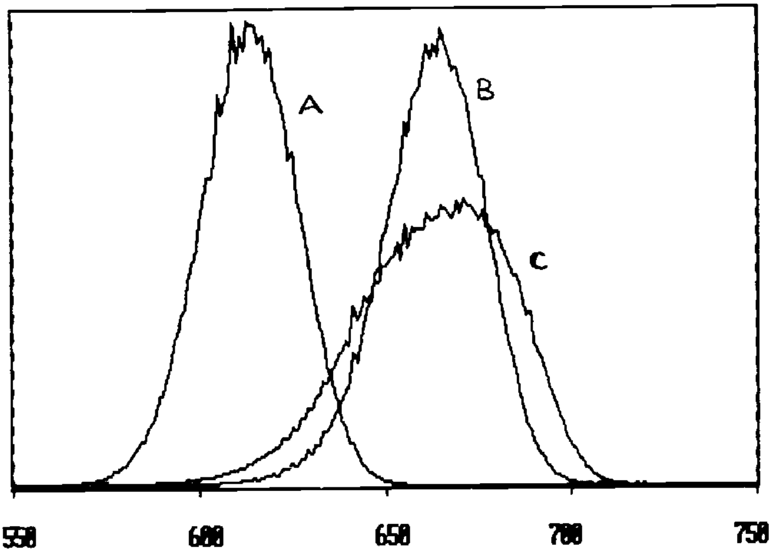
Figure 1. ^{226}Ra spectrum from 2 mL water sample with 18 mL Optiphase Hisafe 3 in 20 mL teflon vial. Alpha emission peaks from left to right are ^{226}Ra (4.78 MeV), ^{222}Rn (5.49 MeV), ^{218}Po (6.00 MeV), and ^{214}Po (7.69 MeV).

In the logarithmic amplitude scale, the alpha peak shape is quite close to being symmetric (Figure 2). At very high alpha particle energies, an asymmetric shape may result due to increased losses close to the vial walls, c.f. ^{214}Po, 7.7 MeV.

PERFORMANCE IN ALPHA COUNTING

Glass vials contain U and Th isotopes, their contribution results in a wide background spectrum under the alpha peaks (Figures 3 and 4). These alpha particles are emitted in a 2 π geometry and are subject to losses due to their need to penetrate through a layer of glass in order to reach the cocktail. Background also contains slow fluorescence from the glass excited by the environmental radiation.

Attenuation of the beta-like decay events among alpha-like ones by pulse shape analyzer is given in Table 1. Typical background count rates and lower



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[A] 1500.000 CPM/ch   89.17 min   A:\NOGUARD\Q011501N.001  SP#12
[B] 2300.000 CPM/ch    2.84 min   A:\CLAM\Q010101N.001  SP#12
[C] 1500.000 CPM/ch   89.17 min   A:\NOGUARD\Q021601N.001  SP#12
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INTEGR( 550- 750)  [A] 47150.711 CPM [B] 73346.578 CPM [C] 47482.875 CPM
BUNCH= 1  * 3S *     60.984 (  0.1 %) 402.513 (  0.7 %)  69.226 (  0.1 %)
```

Figure 2. ^{241}Am alpha emission spectrum (5.5 MeV) in teflon vial (A), etched glass vial (B), and in standard glass vial (C).

limits of detection are given for ^{241}Am (5.5 MeV) and ^{214}Po (7.7 MeV) in two instruments.

CONCLUSIONS

There are several advantages to alpha particle counting with the LS method compared to solid state detection, gridded ion chambers, or the nuclear track method, especially when high energy resolution is not required. The LSC performance does not involve a variable geometrical factor, variable counting efficiency, self absorption, and detector contamination. A counting efficiency of at least 98% with a background of less than 0.1 cpm can easily be achieved. Sample preparation is simple; saving time in low level counting. Sample sizes considerably larger than in solid state alpha spectrometry can be analyzed, and enrichment is a simple procedure. Spectrum analysis allows early recognition of alpha-emitting nuclei in the samples and thus time for decision making if any further analyses are required.

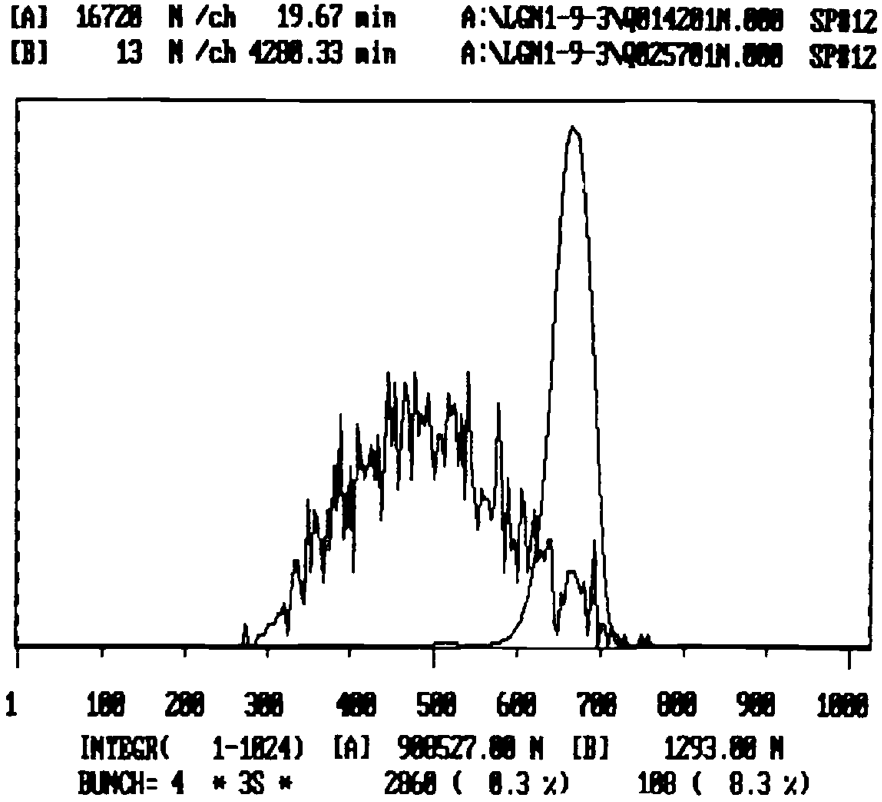
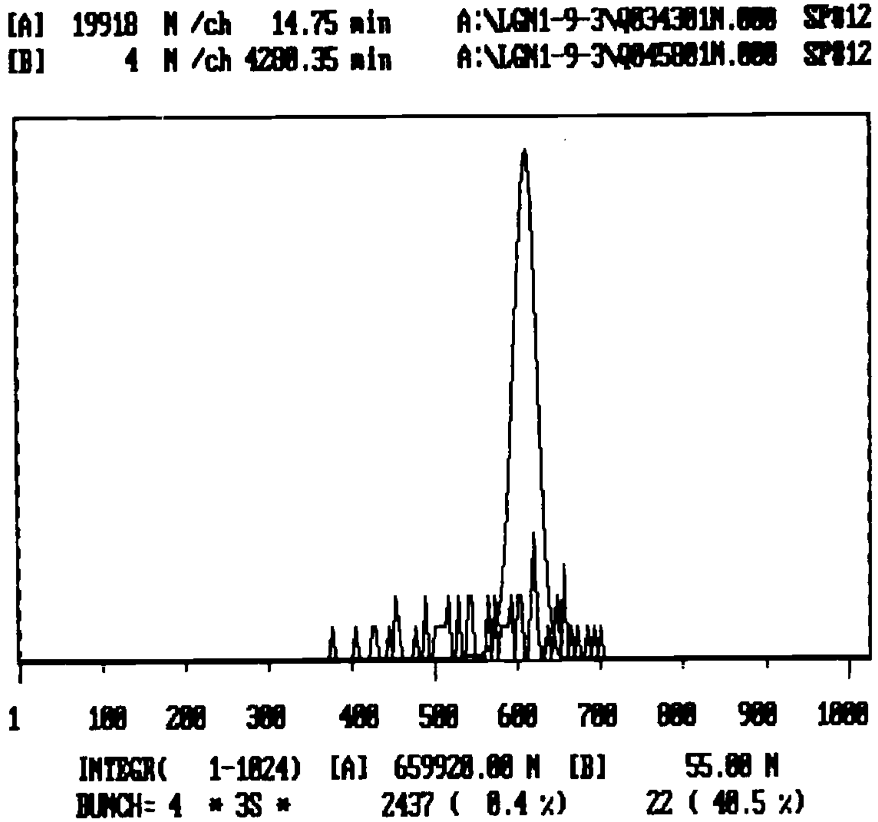


Figure 3. ^{241}Am alpha emission and background spectra in a standard glass vial (1 mL water + 9 mL Lumagel + 20% w/v naphthalene). Background count rate in full window is 0.38 cpm and 0.002 cpm in ^{214}Po window.

Table 2. Alpha Peak Background (cpm) and Lower Limit for Detection (LLD, Bq/L) for 100 min Counting Time in the Wallac 1220 Quantulus and 1219 SM (which contains no guard counter). Sample is 2 mL water + 18 mL Lumagel + 20% w/v Naphthalene in Equilibrium with Air

Instrument	Vial	bkg	LLD	Comment
1219 SM	glass	0.23	1.2	Am-241
1219 SM	glass, op	0.11	0.8	Am-241
1219 SM	glass, op	0.03	0.08	Po-214
1219 SM	teflon	0.02	0.05	Am-241
1220	glass	0.03	0.08	Am-241
1220	teflon	0.005	0.01	Am-241
1220	teflon	0.001	0.002	Po-214



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[A]  19918  N /ch    14.75 min     A:\LGN1-9-3\Q8343801N.000  SP#12
[B]      4  N /ch  4200.35 min     A:\LGN1-9-3\Q8458001N.000  SP#12
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INTEGR(   1-1024)  [A]  659920.00 N  [B]      55.00 N
BUNCH= 4  * 3S *         2437 (  0.4 %)      22 ( 40.5 %)
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Figure 4. ^{241}Am alpha emission and background spectra in a teflon vial (1 mL water + 9 mL Lumagel + 20% w/v naphthalene). Background count rate in full window is 0.013 cpm and 0.001 cpm in ^{214}Po window.

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