Application Note

Alpha/Beta ABA-005

High Throughput Screening of Samples Containing Alpha & Beta Radionuclides: an Overview of Methods

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Introduction

The growing need to quantitate alpha emitting radionuclides in the environment and in nuclear fuel processing and disposal has resulted in dramatic increases in gross alpha/gross beta measurements. In fact, gross alpha/gross beta counting has become the most widely used method of monitoring for the presence of radioactivity. Much of the interest is due to the need to address safety, regulatory compliance, and disposal issues for both alpha and beta radionuclides. The potential for litigation has also heightened interest in screening samples for gross alpha/gross beta activities.

Traditionally, gas flow proportional counting has been used for making gross alpha/gross beta measurements. However, gas flow proportional counting has some disadvantages when compared to liquid scintillation counting (LSC). Counting efficiencies for alphas are 30%, or less, compared to nearly 100% for LSC. Alpha particles, and to a lesser extent beta particles, are attenuated by salts and solids dried onto a planchet. In fact, 100 mg of solids will reduce the absolute alpha counting efficiency from 30% to 10%. Attenuation is not a problem with the LSC method because the sample is completely dissolved in the counting cocktail. No energy resolution is possible with gas flow systems. There is also no way to detect tritiated water with gas flow methods since the sample must be evaporated to dryness, thus volatilizing the tritium.

Recently, Packard Instrument Company has introduced a liquid scintillation counter employing a Pulse Decay Analysis (PDA) technique. This system discriminates alpha events from beta events in samples based on the duration of the light pulses they produce in a liquid scintillator. PDA uses a special pulse decay discriminator to categorize the pulses as either alpha events or beta events and stores the events appropriately in separate multichannel analyzers (MCAs).

Liquid scintillation counting (LSC) with alpha/beta discrimination capability is rapidly becoming the method of choice when screening for gross alpha/gross beta activity. Among the attractive features of LSC are its near 100% counting efficiency for alphas; the reduced sample preparation for most samples, especially liquids; low backgrounds (0.1 CPM in the region of a 5 MeV alpha emitter); some degree of energy resolution; and sample changing capability.

The advantages of using LSC with PDA for alpha/beta discrimination make this technique ideal for high throughput screening of water, air filters, and swipes. The technique may also be applied to soil assays, as well as urine bioassays. LSC with PDA provides a rapid, simple alternative to laborious alpha spectrometry if isotopic specific analysis is not desired. These three methods (LSC utilizing PDA, gas flow proportional counting, and alpha spectrometry) are compared for their applicability to high throughput gross alpha/gross beta sample screening.

A Comparison of Gas Flow Proportional Counting, LSC, and Alpha Spectroscopy Methods

Basically, two methods exist for the simultaneous quantitation of alpha and beta emitting radionuclides in the same sample: gas flow proportional counting and LSC with alpha/beta discrimination. Separation chemistry and high resolution alpha spectroscopy are applied when it is necessary for isotope specific analysis. The first two methods are useful for high throughput screening when rapid sample analysis and quantitation are re-

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quired. The alpha spectrometry method is generally accepted for the determination of alpha emitting nuclides in urine. Alpha spectrometry requires extensive sample preparation to isolate the alpha nuclides of interest from other alphas with similar energy, and to remove beta emitting nuclides. Therefore, alpha spectrometry is not generally suited for high throughput screening.

Gas Flow Proportional Counting

Sample preparation involves evaporation of the sample to dryness on a planchet which concentrates the solid material present. The amount of solid material deposited onto the planchet will severely attenuate the alpha particle. Sometimes it is necessary to include a filtration step if there is a large amount of particulate material in the sample. The presence of even milligram quantities of material will greatly affect the counting efficiency for alpha and, to a lesser extent, beta emitters. A mass attenuation curve and its mathematical equation for the alpha emitter ^{239}Pu is shown in Figure 1. Once the sample is prepared on a planchet, it is placed under the detector.

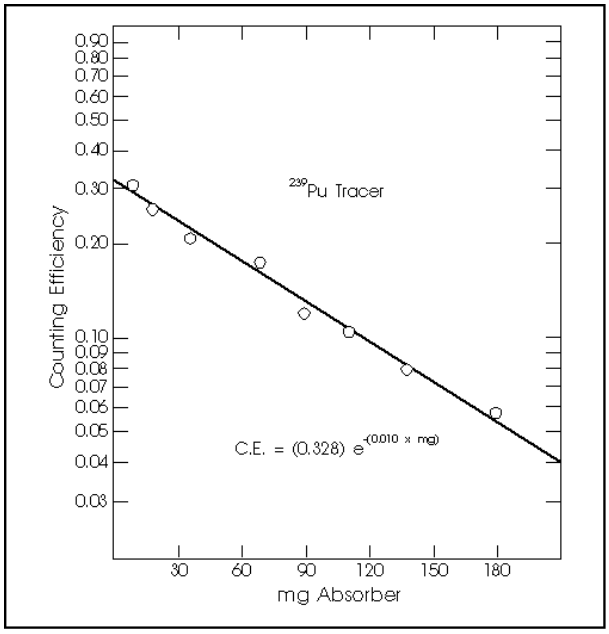

Figure 1.
Mass Attenuation Curve - mg Absorber vs. Counting Efficiency.

A typical gas flow detector consists of a tube with a high conductivity copper body, an anode wire, a thin Mylar window, and gas inlet and outlet ports. The detector is filled with P-10 counting gas (a mixture of argon and methane). Alpha and beta particles penetrate the window and ionize the gas. This ionization results in further ionizations (ionization avalanche) because of the potential applied between the anode and cathode. Ultimately, this avalanche of ionization accumulates on the anode wire and produces an electrical pulse. The pulse amplitude is based on the degree of ionization caused by the incident beta or alpha particle. Cosmic background is eliminated by using a guard detector and an anti-coincidence detection scheme. Separation of the alpha from the beta emitters is on the basis of pulse height. This can be accomplished because the alpha emitters produce a pulse which is about 40 times that of the beta pulse. In modern systems, the pulse signal is amplified, digitized, and sent to a Multichannel Analyzer (MCA) where Regions of Interest (ROI) are established to categorize the alpha and beta events. The system requires proper adjustment of both the high voltage and gain to optimize the discrimination of alpha from beta particles. The misclassification of alpha from betas is 1-3%. Typical counting efficiencies are 30% for alphas and 40% for betas with alpha backgrounds of 0.05-0.5 CPM. No energy resolution can be obtained with gas flow proportional counters.

Alpha/Beta Liquid Scintillation Counting

The second method for simultaneous quantitation of alpha and beta particles in the same sample is liquid scintillation counting using PDA. PDA uses special pulse decay time discrimination electronics to differentiate alpha pulses from beta pulses on the basis of the time it takes for the pulse to decay in a liquid scintillator. A Pulse Decay Discriminator (PDD) is optimized to accurately separate the pulse types and store the events in separate MCAs. Alpha/beta discrimination electronics is necessary because alpha particle energies appear to be approximately
1/10 their kinetic energy when counted in a liquid scintillation cocktail. Therefore, a 5.00 MeV alpha will be detected as a symmetrical peak at approximately 500 keV. In the case where both alpha and high energy beta particles are present in the same sample, they cannot be differentiated on the basis of their energy. This is illustrated with the overlapping spectra of ^{36}Cl and ^{241}Am; see Figure 2. Even though alphas appear to have 1/10 their emitted energy, counting efficiency is maintained at nearly 100% because every decay event is detected even when quenched.

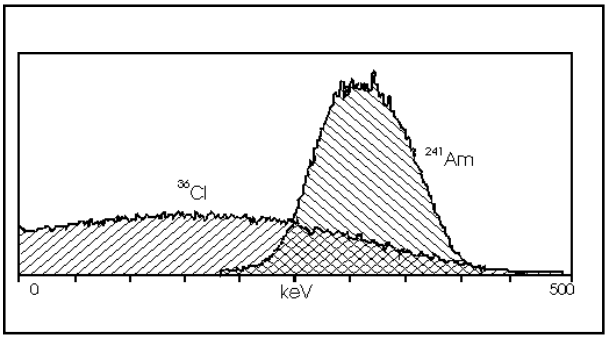

Figure 2.
Multichannel Analyzer Display Alpha/Beta LSC - ^{241}Am and ^{36}Cl.

PDA provides the means to separate alpha events from beta events despite their similar energies in a liquid scintillation counter. This is based on the fact that alpha particles produce light pulses in the cocktail that have a longer duration time than beta pulses. A greater portion of the light produced from alpha pulses comes from excitation which results in the tailing of the initial prompt portion of the alpha pulse. Thus, it takes longer for the light to dissipate for alpha produced pulses than for beta produced pulses. The difference in the duration of alpha and beta pulses is illustrated in Figure 3. This pulse decay time phenomenon can be used to establish an optimum alpha/beta discriminator setting. Only a pure beta and a pure alpha standard are required to establish this optimum setting. A plot of percent spillover of alpha events into the beta MCA and beta events into the alpha MCA is automatically generated by the instrument. The optimum discriminator setting is automatically calculated from the intersection of the two curves. A typical percent spillover plot is shown in Figure 4. Once the optimum setting is determined, it is stored in a library for use in any counting protocol. Under optimum conditions, misclassifications as low as 0.5% can be obtained for routine samples. Energy resolution can be obtained for both the beta and the alpha MCAs. Counting efficiency for alpha emitters is nearly 100% and background for the entire alpha MCA is 0.5-1.0 CPM and 0.1-0.3 CPM for an optimized alpha counting region.

Alpha Spectrometry

The third method which can be used to detect alpha particles is alpha spectrometry. This method is generally accepted for measurement of specific actinides in environmental and bioassay samples and has been applied to direct counting of filters.[1] The detector used is a semiconductor composed of silicon. The advantage of alpha spectrometry is that it provides excellent alpha-alpha energy resolution (15-30 keV) and has very low backgrounds (0.003-0.01 CPM). The disadvantages are that alpha spectrometry has a counting efficiency of about 25% and requires extensive separation chemistry to prepare the samples including sample mounting. Basically, sample preparation involves a series of multiple steps using liquid-liquid extractions or ion-exchange chromatography. Also, the addition of a tracer is necessary to measure chemical recovery. Separation chemistry is critical because improper separation could result in misidentification. Improper mounting will result in poor resolution (smearing). This makes identification of individual isotopes difficult if not impossible. Sample mounting (the sample is either electroplated or precipitated with microgram quantities of some rare earth fluoride) is important because a layer deposit that is too thick will cause self-absorption problems and non-reproducible results. Because of extensive sample preparation, separation chemistry, and sample mounting, alpha spectrometry is usually not used for high throughput screening. However, it is included in the following comparison because it is widely used for alpha measurements.

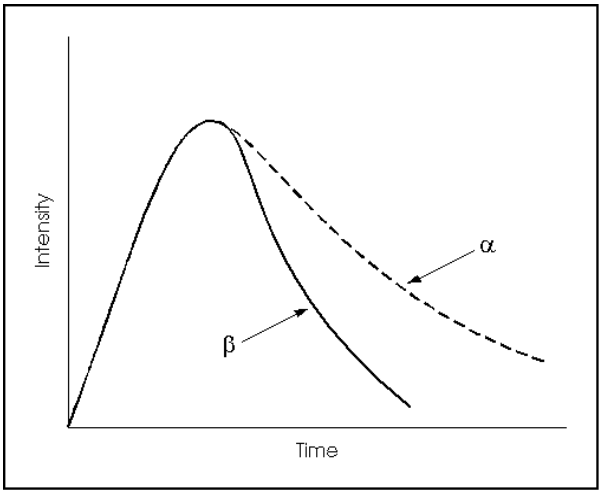

Figure 3.
Characteristic Light Pulse Shapes of Alpha and Beta Pulses in LSC.

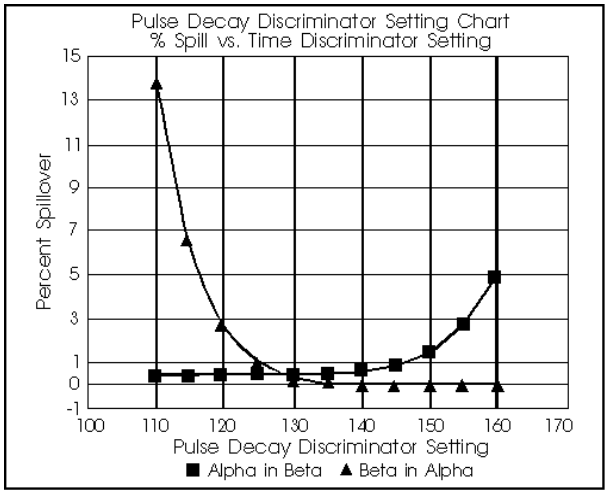

Figure 4.
Typical Alpha/Beta Crossover Plot - ^{241}Am and ^{36}Cl.

Comparison of Performance Parameters

Quantitatively, various counting methods are compared on the basis of Figure of Merit (FOM). The FOM is expressed as the square of the counting efficiency (E^2) divided by the background (B). E^2 is the percent counting efficiency squared and B is the background CPM. To evaluate the sensitivity of these three methods, an additional factor (attenuation) is applied to the FOM calculation. The modified figure of merit calculation then becomes (E^2/B) x Attenuation Factor (AF). Attenuation is caused by sample self-absorption and results in a loss of counting efficiency. Table 1 contains a comparison of the three methods for efficiency, background, and attenuation.

The sensitivity of these methods can be compared by modifying the FOM to include attenuation:

	(E^2/B)(AF)
Gas Flow Proportional Counting	7350
LSC with PDA	36200
Alpha Spectrometry	208333

The background used for the LSC (PDA) FOM was 1.0 CPM. FOM's for gas flow proportional counting and alpha spectrometry were calculated using 0.05 CPM and 0.003 CPM backgrounds respectively. The efficiencies used are reported in Table 1.

From these data, it is quite clear that if high throughput screening of mixed alpha/beta samples is the objective, then the LSC PDA method will provide good sensitivity. Although LSC gives a lower FOM than alpha spectrometry, it will not require all of the extensive sample preparation steps. If high throughput screening is the objective, then initial screening for activity by the LSC method can be followed by alpha spectrometry for the samples that have alpha activity.

Another parameter to compare is Minimal Detectable Activity (MDA). The calculation used here is defined by the equation in Figure 5.

If counting times of 30 and 100 minutes are compared and a sample volume of 10 mL is used, the following MDA's are calculated as shown in Figure 6. The following backgrounds were used in the MDA calculation: LSC, 0.25 CPM; gas flow 0.05 CPM; alpha spectrometry, 0.003 CPM. Attenuation and counting efficiency used are reported in Table 1.

	Alpha Background (CPM)	**Alpha Counting Efficiency**	**Attenuation**
Gas Flow Proportional Counting	0.05-0.5	35%	0.30
LSC (PDA)	0.1-1.0	95%	none
Alpha Spectrometry	0.003-0.01	25%	none (with proper sample preparation)

Table 1.

$$MDA = [2.71 + (4.65\ B^{1/2})] / (2.22 * T * E * AF * V * Y)$$

Where:

B = total counts for an appropriate reagent blank

T = count time in minutes

E = counting efficiency

AF = attenuation factor (if applicable)

V = sample volume (if applicable) in liters

Y = chemical yield (if applicable)

This equation will provide the result in pCi/L.

Figure 5.

These data clearly demonstrate that LSC provides MDAs that are comparable with alpha spectrometry and lower than gas flow proportional counting for equivalent size samples.

Throughput

The most important considerations in high throughput screening assays are the amount of time it takes to prepare samples, count them, and achieve the required MDA. Screening for gross alpha/gross beta activity encompasses various types of samples, including filters (air monitoring or swipe assays), water, solid materials, and urine.

Sample preparation for gas proportional counting requires concentration, evaporation, and solubilization with acid for aqueous type samples. For solid samples, wet or dry ashing is necessary to digest the material.

	pCi/L (Bq/L) 30 minutes	pCi/L (Bq/L) 100 minutes
Gas Flow Proportional Counting	120.2 (4.4)	56.2 (2.1)
LSC (PDA)	24.4 (0.9)	12.3 (0.5)
Alpha Spectrometry	24.7 (0.9)	9.5 (0.4)

Figure 6.
MDA Calculations (Alphas).

The alpha spectrometry method is the most time consuming method for sample processing because chemical separation and/or chromatography, and sample mounting (a thin film) for analysis is required. Urine samples are typically processed with such a scheme.[2] For screening purposes, this method is not practical.

In terms of sample preparation, LSC provides the best means of insuring that the time and steps required are minimal. The basic requirement of any LSC sample preparation scheme is to ensure that the samples are homogeneous (solubilized) and that the samples are soluble in the cocktail. For aqueous samples, direct addition of sample to the cocktail is possible. For solid materials, ashing, elution, or dissolution are methods that are used to prepare samples. Even if ashing or solubilization is required, sample preparation is usually reduced because once the sample is solubilized, it can usually be added directly to the cocktail. Most sample preparation procedures for LSC require less than six hours, and thus the turn around time for samples is less than 24 hours. In addition, Tri-Carb alpha/beta LSC's can count 408 large vials (20 mL) or 720 small vials (4 or 7 mL) automatically. Rapid and simplified sample preparation for the LSC technique is routine. The sample preparation schemes for gas flow counting or alpha spectroscopy are adaptable to LSC and simplified. Because of the high counting efficiencies, LSC can provide sensitivities for a one liter sample similar to, if not greater than, that of alpha spectrometry.

Meeting MDA requirements is necessary because it will determine whether a sample requires further processing. If the gross alpha activity is less than 15 pCi/L, excluding radon and uranium, or the gross beta activity is less than 50 pCi/L, many samples require no additional analysis because the sample is in compliance. These guide values are recommended in the EPAs National Interim Primary Drinking Water Regulations. These MDAs are easily achievable with the LSC PDA method. For example, with an unconcentrated 10 mL water sample counted for 60 minutes, an alpha MDA of 16.4 pCi/L is achievable (95% counting efficiency, 0.25 CPM optimized background, 95.5% confidence). This is compared to approximately 400 minutes of counting time that is required for gas flow (35% counting efficiency, 0.5 attenuation factor, 0.05 CPM background). The difference may even be greater between the two methods since the gas flow attenuation factor applied is conservative.

The time required to achieve the required MDA is an important factor for urine bioassay, since U.S. DOE Order 5480.11 suggests utilization of the detection limits specified in ANSI N13.30 of 0.1 pCi/L for uranium and 0.06 pCi/L for plutonium. An alpha MDA of 0.12 pCi/L can be obtained with the LSC PDA method (95% counting efficiency; 0.25 CPM background) compared to 0.09 pCi/L with alpha spectrometry (25% counting efficiency; 0.003 CPM background) for a 100 minute count time and a starting volume of 1 L. The LSC PDA method can achieve a comparable MDA for samples processed similarly and supports the decision to perform additional analysis by high resolution alpha spectrometry.

Summary of the Three Methods

Table 2 provides a review of the three methods on several issues.

With the LSC method, up to 10 mL of sample can be counted directly or after processing by separation chemistry, concentration, or precipitation. No matter how the sample is prepared, the LSC PDA method is a very sensitive method of analysis because of nearly 100% counting efficiency and low backgrounds in the alpha MCA (<1.0 CPM for the entire MCA).
The sample changing feature of a Packard Tri-Carb® TR/AB alpha/beta LSC provides the capability for unattended counting of 720, 7 mL samples or 408, 20 mL samples. Certain gas proportional counters offer sample changing capacities of up to 240 samples. The alpha spectrometry method analyzes one sample at a time with no sample changing capability. Therefore, the LSC method provides the maximum unattended sample processing capacity.

Automatic protocol selection with the Packard Tri-Carb TR/ABs provides the versatility to count a variety of different applications unattended by using a separate protocol for each. For example, it is possible to dedicate protocols to counting conditions required for specific samples such as water, air, urine, and soil.

There are many reasons for making gross alpha/gross beta measurements. Gross alpha/gross beta measurements will provide sufficient information to conclude that no additional analyses are necessary, depending on the sample. Sample screening is a means of rapidly determining if the samples are in compliance with NRC, EPA, or DOE regulatory activity action levels. When monitoring the work environment, higher throughput leads to faster answers and prompt corrective action when required.

	LSC	Gas Proportional	Alpha Spectroscopy
MDA (pCi/L) (parameters used from Fig. 6; 100 minute count time calculation)	12.3	56.2	9.5
Sample capacity	408-720	</=240	1 per detector
Automatic protocol selection	yes	no	no
Tritium quantitation	yes	no	no
Energy discrimination	yes	no	yes (alpha)
% misclassification	</=0.5 (optimum)	1-3	separated

Table 2.
Comparison of Three Methods for Alpha Screening.

An additional benefit of LSC is its ability to quantitate tritium, especially in water and surface monitoring samples. The alpha spectrometry method is not able to quantitate tritium. The gas proportional counting method cannot quantitate tritium unless the counting window is removed. Even then, counting efficiency is extremely low and contamination of the detector is a concern. LSC/PDA provides the ability to detect tritium from surface swipes and water, while simultaneously quantitating alpha activity. In fact, MDAs for tritium in a low level count mode of less than 10 T.U./Liter (71.9 DPM/L) are possible by direct sample addition.
Alpha spectrometry provides 15-30 keV of energy resolution for alphas, but no alpha/beta discrimination. Gas proportional counting provides only alpha/beta discrimination but no energy resolution of either alpha or beta particles.

LSC/PDA provides both alpha/beta discrimination and energy resolution for both alphas and betas. The energy resolution of alphas is 400-500 keV FWHM, and energy resolution for betas is unchanged from normal LSC.

An important issue in alpha/beta sample screening is the degree of misclassification of alpha and beta emitting radionuclide activities. Alpha spectrometry uses separation techniques to physically separate the alpha and beta emitting radionuclides. The gas proportional counter can easily distinguish alphas from betas with 3% or less alpha crosstalk into the beta channel and 1% or less beta crosstalk into the alpha channel. Under optimum conditions, the LSC/PDA method can provide 0.5% or less crosstalk. This can be important if the ratio of beta to alpha in the sample is high. Up to a 200 fold excess of beta to alpha activity can be assayed for alpha activity by direct sample addition with the LSC method.[3] Accurate alpha activity quantitation, with up to a 10,000 fold excess of beta to alpha activity, is possible with the use of extractive scintillators.[4]

EPA Recognition

Liquid scintillation counting has been approved as an alpha counting method by the EPA as shown in the following quote from the EPA 40 CFR Parts 141 and 142, National Primary Drinking Water Regulations. Proposed rule that appeared in the Federal Register (Vol. 56, No. 138/page 33093, Thursday, July 18, 1991):

"Counting methods. Alpha Emitting Radionuclides (Gross alpha particle activity, Radium-226, Radon-222, and uranium)-Alpha counting Methods-Alpha particles are characterized by an intense loss of energy in passing through matter. This intense loss of energy is used in differentiating alpha radioactivity from other types by dense ionization or intense scintillation it produces.

Alpha counting methods, which measure alpha radioactivity, are applicable in determination of gross alpha particle activity, Radium-226, Radon-222, and uranium. Alpha radioactivity can be measured, after various sample preparations, by one of several types of detectors in combination with appropriate electronic components. The techniques for measuring the alpha emitters use gas flow proportional counters, scintillation cell systems and liquid scintillation counters, in conjunction with electronic components such as high voltage power supplies, preamplifiers, amplifiers, scalers and recording devices. Additional techniques using fluorometry and alpha spectrophotometric techniques are being proposed for uranium.

Direct low volume liquid scintillation counting of alpha emitters with a commercially available instrument is also employed in the proposed methods. A liquid scintillator or organic phosphor is combined in an appropriate mineral oil or organic base scintillator 'cocktail' with the water sample. Mixing achieves a uniform dispersion before counting. This replaces the planchet or disk preparation that occurs before the counting step in the scintillation technique."

Conclusion

From the comparison of the issues in Table 2, it is clear that the LSC method is extremely sensitive for measuring alpha activity with some energy resolution. A large number of samples (408-720) can be quantitated using automatic protocol selection for different applications. These benefits are the reasons that the LSC PDA method is becoming the preferred method for gross alpha/gross beta screening.

References

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