Application Note

Alpha/Beta ABA-003

The Effect of Quench on Quantitating Alpha Radionuclides by Liquid Scintillation Counting

Abstract

Detecting and quantitating alpha emitting radionuclides are routine tasks in nuclear energy and environmental monitoring. For this purpose, liquid scintillation counting (LSC) is an excellent method because alphas are counted with nearly 100% counting efficiency with low backgrounds, even with severe quenching. Quenching agents such as inorganic acids, organic acids, and extractive scintillators are often required to prepare samples when assaying high-level radioactive waste. Spectral data show that in spite of a shift to lower energy and a general broadening of the spectra, 100% counting efficiency is maintained for alpha nuclides. Extractive scintillators show improvement in alpha resolution compared to cocktails containing emulsifiers However, resolution generally degrades with an increase in quench. This effect of quench on counting efficiency and resolution for alpha emitters is discussed.

Introduction

With increased concern about radionuclides in the environment, it has become increasingly important to accurately quantitate alpha emitters. Nuclear fuel fabrication and processing, nuclear reactor operation, radioactive ore mining operations, waste storage facilities, and environmental monitoring laboratories detect and quantitate alpha activity. Liquid scintillation is often the method of choice when high throughput screening for the presence of alpha activity is required. In fact, recently the U.S Environmental Protection Agency (E.P.A.) has validated the method (E.P.A. 40 CFR Parts 141 and 142, National Primary Drinking Water Regulations) for the measurement of gross alpha activity for ^{226}Ra, ^{222}Rn, and Uranium in drinking water. Liquid scintillation has recently been applied to the assay of transuranium elements in high-level radioactive waste.[8] This study explores the effect of quench on alpha counting efficiency and resolution.

Alpha Particle Detection by Liquid Scintillation

In order to understand the effect quenching has on alpha particle detection, a brief introduction of counting alpha particles by liquid scintillation is necessary.

The liquid scintillation detection process involves converting the kinetic energy of the alpha particle into light flashes. The intensity of the light emission and energy of these alpha particles are directly proportional to one another.

Most alpha particles are emitted with kinetic energies in the range of 2-8 MeV. However, much of the energy is not directly converted to light but is dissipated through molecular ionization. This ionization, coupled with molecular damage from decomposition and free radical formation, gives rise to a short-lived process called "ionization quench."[1,2] Because of this phenomena, a 5 MeV alpha particle will appear to have the energy of 500 keV or about one tenth of its particle energy. Therefore, all alpha-emitting radionuclides, are detected in the range of 200-800 keV. This makes detection and quantitation possible by conventional liquid scintillation counting.

Alpha emitting radionuclides, when quantitated by liquid scintillation counting, generally produce a symmetrical peak around the energy maximum of the alpha particle (1/10 its actual energy). The energy spectrum for beta emitting radionuclides is different from alpha particles when detected in a liquid scintillation counter because beta emitters produce a continuous spectra from zero to the energy maximum (E_{max}) for the radionuclide.

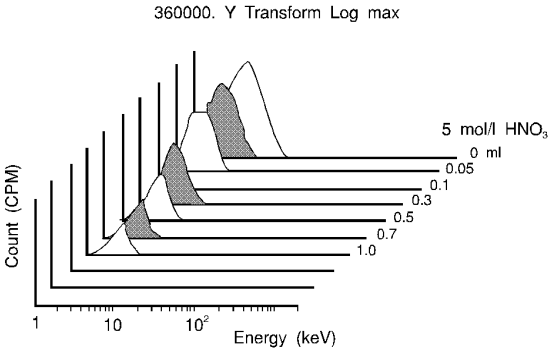
Quenching Effects on Alpha Particle Detection

The effect of quenching on both beta and alpha emitting radionuclides is illustrated in Figures 1 and 2. Figure 1 is a three-dimensional spectral plot of seven ^{3}H samples

Packard A Packard BioScience Company

Packard Instrument Company 800 Research Parkway Meriden, CT 06450
Tel: 203-639-2598 1-800-323-1891 Fax: 203-639-2172
Web site: http://www.packardinstrument.com E-mail: webmaster@packardinstrument.com

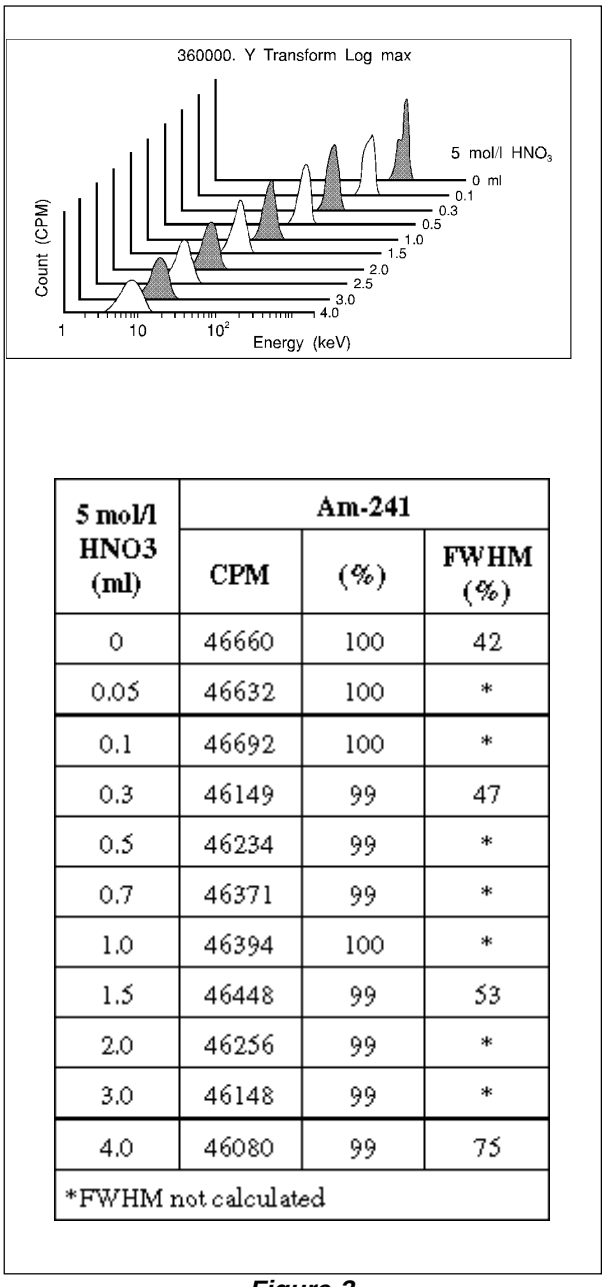
that have been quenched with 0-1.0 ml of 5 M nitric acid in 10 ml of cocktail (5 g/L PPO, 0.5 g/L dimethyl POPOP, 50 g/L naphthalene, toluene and triton X-100). As can be seen in Figure 1, the area under each peak as well as the energy or pulse-height spectrum for tritium is substantially reduced with an increase in acid loading. This is the result of chemical quenching that absorbs some of the energy of the beta particle before and during the conversion of the beta particle energy to flashes of light. The inserted table in Figure 1 shows the counting efficiency as a percentage relative to a sample containing no quenching agent. The relative counting efficiency (number of light flashes - CPM) and the energy (intensity) is decreased to 13% when 1.0 mL of 5 M nitric acid is added to the sample.



5 mol/l HNO4 (ml)	H-3	
	CPM	(%)
0	126287	100
0.05	115834	92
0.1	102218	81
0.3	61211	48
0.5	39846	32
0.7	25239	20
1.0	16091	13

Figure 1.
Quenching Effects of 5 M HNO_3 on ^{3}H.

In contrast, for ^{241}Am, an alpha emitter, acid loading does not change the counting efficiency, even with the addition of up to 4.0 mL of 5 M nitric acid in the same cocktail. Figure 2 illustrates how the peaks are simply shifted to a lower energy and broadened as the amount of acid is increased. This phenomenon is generally observed when quenching alpha particles. Quenching affects the intensity at which the alpha peak is observed, but not the counting efficiency. Even with the addition of 4.0 mL of 5 M nitric acid, the counting efficiency remains at 100%. This is significant since samples for alpha counting typically contain strong acids.

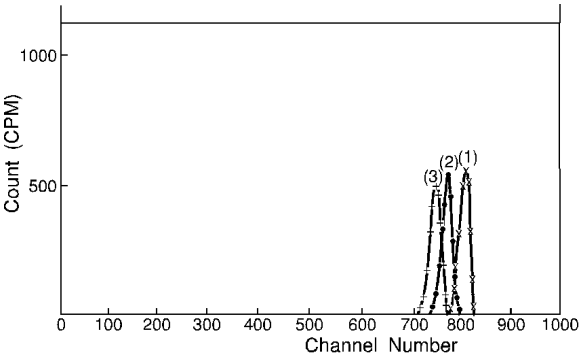


5 mol/l HNO3 (ml)	Am-241		
	CPM	(%)	FWHM (%)
0	46660	100	42
0.05	46632	100	*
0.1	46692	100	*
0.3	46149	99	47
0.5	46234	99	*
0.7	46371	99	*
1.0	46394	100	*
1.5	46448	99	53
2.0	46256	99	*
3.0	46148	99	*
4.0	46080	99	75
*FWHM not calculated			

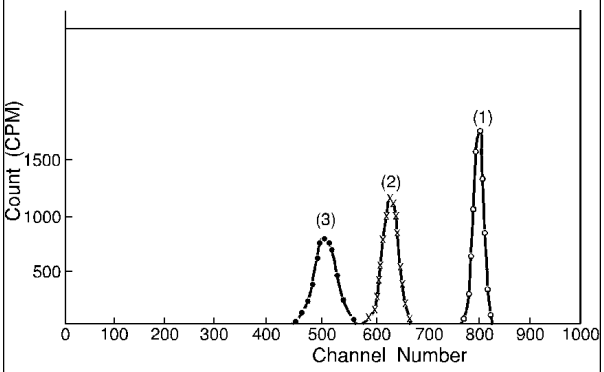
Figure 2.
Quenching Effects of 5 M HNO_3 on ^{241}Am.

A study has been performed to investigate the effects of quenching caused by adding various amounts of extractive agents to cocktail for the assay of transuranium elements in high-level radioactive waste.[8] The relationship between ^{239}Pu counting efficiency and the concentration of two extractants, trioctylphosphine oxide (TOPO) and

triisoctylamine (TiOA), in a toluene 5 g/L PPO cocktail is shown in Figure 3.



TOPO Extractant (mol/l)	Counting Efficiency (%)	FWHM (%)
(1) 0.0	100	4
(2) 0.5	98	4
(3) 1.0	99	4



TiOA Extractant (mol/l)	Counting Efficiency (%)	FWHM (%)
(1) 0.0	100	4
(2) 0.5	99	7
(3) 1.0	100	13

Figure 3.
Quenching Effects of TOPO and TiOA extractants on ^{239}Pu 10 mL 5 g/L PPO-toluene scintillator.

The data show that there is virtually no loss of counting efficiency due to the addition of these two extractants. In fact, this is generally the case with most extractive agents used in this study. More information on the quench effect of various extractants is available in Packard Alpha/Beta Application Note ABA-002. Although the addition of extractive agents will shift the alpha peak to a lower energy, similar to the effect described for the addition of inorganic acid (see Figure 2), the counting efficiency is not changed at any quench level. At the same time, background in this study was found to be about 0.9 CPM for the entire alpha MCA from 0-2,000 keV, and approximately 0.1 CPM in the region of 5 MeV alpha emitters. This translates into an MDA of 0.5 mBq/10 mL (1.35 pCi/L, 1000 minute count time, at the 3 Sigma confidence level).

Alpha Spectrum Analysis and Resolution

Typical alpha energy resolution in LSC is approximately 25% Full Width at Half Maximum (FWHM)*. [3-6] Increased quenching will reduce the pulse-height (energy) of the peak and degrade resolution. The extent of the degradation is dependent on the nature of the quenching agent and the cocktail. For example, chemical quenchers, with properties similar to carbon tetrachloride (CCl_4), have been shown to dramatically reduce the pulse-height spectrum of ^{210}Po when counted in 10 mL of toluene/10 g/L BBD scintillator, but do not seriously degrade resolution while quenching InstaGel™ (Packard Instrument Company, Inc.) with water did not seriously affect either the pulse-height or resolution.[7] Color quenchers have been shown to reduce the pulse heights by absorption of the light and they degrade resolution.[7] Inorganic acids, such as nitric acid, cause a shift in the alpha peak to a lower energy, but do not cause a loss of counting efficiency (see Figure 2). With little or no acid added to the cocktail, the resolution is about 40% FWHM. However, with increasing volumes of 5 M acid, resolution degrades to 75%.

During the isolation of alpha emitting radionuclides in nuclear fuel processing and other related processes, organic extraction techniques are employed to separate alpha emitting nuclides from one another, as well as from contaminating beta nuclides. The extractant is added to the aqueous sample, the acid concentration is adjusted, and the alpha emitting radionuclides are selectively extracted into the organic phase which contains a primary and secondary scintillator. During this process two phases are formed, the organic phase is removed and placed into a scintillation vial for quantitation. Using alpha-selective extractive scintillators generally improves resolution. However, the degree of quenching and resolution depends on the type of extractant used and the amount added. Figure 3 shows the results of adding incremental amounts of two types of extractive agents, trioctylphosphine oxide (TOPO) and triisoctylamine (TiOA), which are used to extract ^{239}Pu from high-level radioactive waste.[8] Figure 3 shows that TOPO does not seriously affect the quench of the

sample. With TOPO, there is an insignificant reduction in the pulse-height spectrum while resolution is about 4% regardless of the amount of extractive agent added to the cocktail. In contrast, TiOA shifts the pulse-height spectrum to a lower energy and degrades the resolution to approximately 13%. Most organo-phosphorus extractants like TOPO have little quenching effects, while alkyl amine extractants like TiOA have some effect.[8]

Conclusion

Liquid scintillation counting may be used to quantitate alpha particles for a wide range of applications, such as environmental monitoring, nuclear fuel processing, high level waste management, etc. Liquid scintillation counting of alpha particles provides high counting efficiency (near 100%), which is virtually independent of quench. Low backgrounds are obtained by optimizing the counting region, since alphas are detected in a very defined energy range. MDAs as low as 0.5 mBq/10 mL sample have been reported using extractive scintillators.

Quenching adversely affects alpha resolution. Optimum resolution is obtained by minimizing the quenching of the sample, especially in emulsifier based cocktails. Further improvements of resolution can be achieved by using extractive scintillators. LSC facilitates easier sample preparation for most samples, and provides high counting efficiencies and low background to make it the most sensitive method available to detect alpha radionuclides. LSC is ideally suited to high-throughput screening of alphas in environmental and nuclear facilities.

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