

LIQUID-SCINTILLATION ALPHA-DETECTION TECHNIQUES *

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LIQUID-SCINTILLATION ALPHA-SPECTROMETRY TECHNIQUES

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ABSTRACT

Accurate, quantitative determinations of alpha-emitting nuclides by conventional plate-counting methods are difficult because of sample self-absorption problems in counting and because of non-reproducible losses in conventional sample separation methods. Liquid scintillation alpha spectrometry offers an attractive alternative with no sample self-absorption or geometry problems and with 100% counting efficiency. Sample preparation may include extraction of the alpha emitter of interest by a specific organic-phase-soluble compound directly into the liquid scintillation counting medium. Detection electronics use energy and pulse-shape discrimination to yield alpha spectra without beta and gamma background interference. Specific procedures have been developed for gross alpha, uranium, plutonium, thorium, and polonium assay. Possibilities for a large number of other applications exist. Accuracy and reproducibility are typically in the 1% range. Backgrounds on the order of 0.01 cpm are readily achievable. The paper will present an overview of liquid scintillation alpha counting techniques and some of the results achieved for specific applications.

INTRODUCTION

Counting of beta and alpha emitters shares the problem of sample self-absorption since both types of radiation are relatively non-penetrating. Liquid scintillation has emerged as the premiere method for counting low energy beta emitters. In the life sciences areas, where tritium and ^{14}C radiotracers are used extensively, advanced commercial systems have been developed that provide automatic quench correction schemes and microprocessors to analyze the data at the push of a button. No such development of liquid scintillation equipment for alpha counting and spectrometry has occurred. There appears to be little incentive for vendors of beta liquid scintillation equipment to explore applications to alpha detection. Furthermore, there appears to be a reluctance among others involved in radiation detection to move away from the existing, "traditional", methods and pursue new, possibly improved, concepts. For example, beta liquid scintillation systems currently available have considerable potential for assay of alpha particles, but these opportunities have been neither fully explored or exploited. But of more pertinence to this paper, recent developments in electronic pulse-shape discrimination and in improved solvent extraction techniques provide considerable potential for assaying a variety of alpha-emitting nuclides with up to 100% chemical recovery and 100% counting efficiency.

At the present state of development, alpha liquid scintillation spectrometry allows much more accurate quantification of alpha radiation than is possible by other methods, but does not provide the alpha energy resolution possible under optimum conditions using plate methods. The applicability in any radiation measurement situation must be determined by the requirements of that situation. This paper provides a brief overview of the current status of alpha liquid scintillation counting, its applications and future potential.

DETECTION TECHNIQUES AND APPLICATIONS

In alpha liquid scintillation counting and spectrometry, the nuclide is placed in homogeneous solution in a toluene- or xylene-based solution (scintillation cocktail) containing a fluor (substance that emits light when excited by radiation) and usually

an energy transfer agent such as naphthalene. Scintillation cocktails designed for beta counting generally contain chemicals that allow aqueous samples to be incorporated into them; those designed for alpha spectrometry usually contain an extractant (water insoluble phase-transfer agent) and are designed to solvent extract the nuclide-of-interest from an appropriate aqueous phase. The resulting scintillator/sample is placed in front of a multiplier phototube that converts the light pulses produced in the scintillator to electrical pulses. An appropriate arrangement for alpha liquid scintillation spectrometry is shown in Fig 1. Dual phototubes are not needed, because the thermal electron noise in the phototubes is always well below the alpha-pulse level. The geometry and reflector shape are fairly critical, however, if good energy and pulse-shape resolution are desired.⁽¹⁾

When ionizing radiation deposits energy in a liquid scintillator, most of the dissipated energy is lost via non-radiative thermal relaxation, but a portion of the excited molecules emit light in the 200 to 400 nm range. For alpha or beta radiation the light pulse is proportional to the energy decay deposited in the medium over at least the range 4 to 7 MeV, but differences in specific ionization between alpha and beta particles produce different fluorescence conversion efficiencies and characteristic light pulse shapes. Energy absorbed from beta and gamma radiation is about 10 times more effective in producing light than is radiation absorbed from alpha particles. This difference in energy conversion causes troublesome background interference from beta and gamma emitting nuclides and from external radiation when alpha counting is attempted using beta liquid scintillation equipment. With appropriate scintillators, alpha-produced pulses are approximately 30 nanoseconds longer than are beta-produced pulses. This difference allows beta-produced pulses to be electronically rejected by electronic timing techniques called pulse-shape discrimination (PSD).

Variable quenching is another problem that has been troublesome in alpha liquid scintillation spectrometry. Colored substances and various chemicals may degrade, or quench, the height and shape of the pulses. Thus the use of aqueous-accepting scintillation cocktails, as in beta liquid scintillation counting, and the introduction of nuclides as aqueous samples result in samples with highly variable light-production efficiencies. The use of aqueous-immiscible scintillators into which the nuclides are introduced as organic-phase-soluble complexes can largely alleviate this problem.

ALPHA COUNTING WITH A BETA LIQUID SCINTILLATION SPECTROMETER

Under some conditions, effective alpha counting can be done using a commercial beta liquid scintillation spectrometer and the aqueous-accepting scintillators available for use in beta counting. Useful applications include research activities using specific alpha radionuclides or performing health physics surveys where swipe information is needed immediately to confirm the presence of contamination. Environmental specimens and bioassays may be screened for the presence of radioactivity before committing to additional, more detailed assay. Filter media, particularly glass-fiber filter materials, may be immersed in the scintillator and counted without self-absorption corrections.

The pulse-height spectra for several non-quenched beta emitters and an alpha emitter obtained using a beta liquid scintillation spectrometer are shown in Fig. 2. The detector was coupled to a multichannel analyzer (MCA) for the experiment. The problem of beta interference with alpha counting is illustrated here. As previously mentioned, the fluorescence conversion efficiency for the alpha emitter is about 1/10 that obtained by beta decay (i.e., a 5 MeV alpha particle produces about the same quantity of light as a 0.5 MeV beta particle). However, liquid scintillation methods permit effective 4π detector geometry with detection efficiencies of 100% for all alpha emitters. Color and chemical quenching shift signals toward the noise region and this will reduce the detection efficiency for beta emitters, but considerable quenching is needed before alpha efficiency is affected. Energy resolution using commercial beta liquid scintillation systems is about 25%; so alpha spectrometry is difficult if not impossible to achieve. Depending on the energy width of the alpha discrimination window and the amount of detector shielding, backgrounds in the alpha particle detection region are between 2 and 10 cpm. Spectral stripping of gross alpha from gross beta-gamma activity may be possible for alpha spectra obtained in this way, but there is no mechanism to alleviate the interference of beta- and gamma-produced pulses with the alpha peak. Interferences produced by gamma radiation are similar to those produced by beta radiation, but overall detection efficiency for gammas is considerably lower.

USING ALPHA LIQUID SCINTILLATION EQUIPMENT

Many of the problems associated with alpha counting and spectrometry by liquid scintillation methods, some of which are mentioned above, may be solved by the use of scintillators, detectors and electronics designed for alpha liquid scintillation. The use of an aqueous-immiscible scintillation cocktail containing an extractant or phase-transfer agent suitable for the nuclide or nuclides-of-interest allows the transfer of the alpha emitter(s) and little else to the scintillator. The resulting scintillator/sample is of constant composition and response to radiation and thus produces pulses reproducible in pulse-height and pulse-shape. Such a scintillator/sample may be placed in a detector such as that shown in Fig. 1, and when connected to preamplifier, amplifier, and MCA electronics of the type used for sodium iodide - gamma spectrometry, reproducible spectra with energy resolution of 200 KeV (for 4 MeV alphas) to 300 KeV (for 7 MeV alphas) can be obtained. Typical energy spectra and a comparison with the resolution obtainable with a commercial beta liquid scintillation spectrometer may be seen in Fig. 3.

Some of the extractants that are useful for phase-transferring alpha-emitting nuclides to aqueous-immiscible scintillation cocktails are: bis-(2-ethylhexyl)phosphoric acid, trioctylphosphine oxide, 1-nonyldecylamine (or similar branched primary amines) and trioctylamine (or similar high-molecular weight tertiary amines). Sulfate and perchlorate are suitable aqueous anion systems from which to extract into the alkyl phosphoric acid and sulfate is probably the most useful system from which to extract with amines. The phosphine oxide extracts from nitrate systems, but aqueous systems containing high concentrations of nitric or hydrochloric acids should be avoided because the extracted acids will quench the scintillator severely. Normal principles of solvent extraction should be observed.⁽²⁾ Considerable selectivity as to the nuclide or group of nuclides extracted is possible by choice of extractant, thus aiding in nuclide identification.

Pulse-shape discrimination (PSD) methods may be used to separate alpha from beta decay events if detectors such as that shown in Fig. 1 and an arrangement like that in Fig. 4 are used. PSD has been widely applied to neutron spectrometry and, during the past decade the concept has been adapted to liquid scintillation alpha particle detection.⁽³⁻⁵⁾ Figure 5 shows an application of pulse-shape discrimination to polonium alpha counting. The combination of detector and electronics

for alpha liquid scintillation spectrometry with pulse-shape discrimination is called "PERALS spectrometry" for Photon Electron Rejecting Alpha Liquid Scintillation spectrometry.

PSD electronics may be assembled from commercial components⁽⁶⁾ or a simple, compact unit designed at Oak Ridge National Laboratory may be obtained from their Instrumentation and Controls Division but pulse shape discrimination is successful only if used with a detector and electronics designed for alpha liquid scintillation spectrometry. Commercial beta liquid scintillation equipment introduces pulse distortion via phototube base, preamplifier and amplifier that prevents the use of PSD equipment.

Portable, single-phototube field units have been designed for specific uses. The units include PSD for beta and gamma rejection and provide coincidence MCA gating signals along with energy pulse-height information.⁽⁴⁾ Fig. 5 shows pulse height, pulse shape, and coincidence-gated MCA spectra produced by alpha-emitting radon and progeny in a beta/gamma background. Samples must be deoxygenated for effective PSD. This is easily accomplished by sparging the scintillator/sample for 1 to 2 min with argon or nitrogen gas. Color quenching or chemical quenching will also degrade pulse-shape discrimination; but even without pulse-shape discrimination, alpha quantification and identification are often possible. The amount of beta-gamma background depends on the ambient radiation and on the amount of beta-gamma emitters extracted with the sample if pulse-shape discrimination is not used. With pulse-shape discrimination greater than 99.95% of the beta and gamma pulses can be rejected without loss of any alpha pulses, and background counts of less than 0.02 cpm under a 5 MeV alpha peak are easily attained.

The detection concept coupled with solvent extraction separation techniques has been applied to the assay of uranium and thorium concentrations in phosphate fertilizers,^(8,9) analysis of ^{210}Po in a uranium mill circuit^(10,11) and determination of plutonium and the transplutonium actinides in a variety of sample types. References will be found in a recent review,⁽¹²⁾ and updated procedures⁽¹³⁾ are soon to be published.

CONCLUSIONS

The applications of liquid scintillation detection may be readily extended to perform alpha particle assay of numerous sample types. The accuracy and reproducibility of the determination is almost always better than with plate-counting methods. Commercially available beta liquid scintillation detectors may be used, but the investigator must recognize the problem of interference from beta and gamma radiation. The use of equipment designed for alpha liquid scintillation along with appropriate pulse-shape discrimination equipment and extractive scintillators will allow the collection of useful energy spectra, the rejection of nearly all beta and gamma interference, and in most cases improvement of the accuracy of the determination. Sample concentration by solvent extraction, 100% counting efficiency and low virtual backgrounds may also combine to give lower detection thresholds than in other methods. The opportunity to make significant improvements over existing detection techniques and the increasing applications and demands for rapid, accurate low-level radiation monitoring provide sufficient incentive to explore the liquid scintillation concept further.

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FIGURE CAPTIONS

Fig 1. A Cross Section of a Reflector Phototube Arrangement Suitable for Alpha Liquid Scintillation Spectrometry

Fig 2. Liquid Scintillation Energy Spectra Obtained Using a Commercial Beta Liquid Scintillation Spectrometer and an Aqueous-Accepting Scintillator.

Fig 3. Alpha Energy Spectra; (a) Thorium and Daughters Taken with an Alpha Liquid Scintillation Spectrometer, (b) Thorium-232 and Plutonium-239 Taken with (a) a Beta Liquid Scintillation Spectrometer and (b) an Alpha Liquid Scintillation Spectrometer

Fig 4. A Block Diagram of the Electronics Required for Alpha Liquid Scintillation Spectrometry with Pulse Shape Discrimination to Eliminate the Beta and Gamma Background

Fig 5. An Application of Pulse-Shape Discrimination to Low-Level Polonium Counting Figure(a) shows a spectrum of 1 dpm of polonium-210 with a high background of beta and gamma radiation. In figure (b) the PSD unit is placed in operation; but the count rate is the same as in (a), 8 min. In figure (c) the counting time is 88 hours. The amount of polonium present is calculated to be 0.9 dpm.

Fig 6. Radon and Progeny; Top, Time spectrum; Middle, Alpha Spectrum with Beta and Gamma Background; Bottom, Alpha Spectrum with Beta and Gamma Contribution Removed by PSD.