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**ALPHA LIQUID SCINTILLATION COUNTING:
PAST, PRESENT, AND FUTURE¹**

W. J. McDowell

Chemical Technology Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

MASTER

Beta liquid scintillation counting has been used for about 30 years, and its effectiveness for alpha particles has been known for almost that long; however, the technique has not been widely applied to alpha particle detection because of poor energy resolution, high background, and variable interference from beta and gamma radiation. Beginning with the work of Horrocks in the early 1960s, improvements in energy resolution and background rejection have been made. Further developments at Oak Ridge National Laboratory over the past 10 to 12 years have resulted in improved methods of sample preparation (using liquid-liquid extraction methods to isolate the sample and introduce it into the scintillator) and better instrumentation, including electronic rejection of beta and gamma pulses. Energy resolutions of 200- to 300-keV FWHM and background counts of 0.01 cpm are now routine. Alpha liquid scintillation spectrometry is now suitable for a wide range of applications, from the accurate quantitative determination of relatively large amounts of known nuclides in laboratory-generated samples to the detection and identification of very small, subpicocurie amounts of alpha emitters in environmental-type samples. Suitable nuclide separation procedures, sample preparation methods, and instrument configurations are outlined for a variety of analyses.

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I. BACKGROUND

The fact that liquid scintillation is applicable to alpha counting has been known almost as long as the method has been used for beta counting. According to Horrocks (1974a), the use of aromatic solvents containing dissolved solutes as scintillation detectors for nuclear radiation was first demonstrated by Professor H. P. Kallmann in 1947. Three years later M. Ageno and co-workers (1950) reported counting alpha as well as beta and gamma radiations in xylene solutions of naphthalene, and in 1954 Basson and Steyn (1954) further demonstrated the usefulness of liquid scintillation for alpha counting. The light output of a liquid scintillator has also been shown to be directly proportional to alpha energy to a useful degree over the range 4 to 7 MeV, an energy range that includes the alpha energies of the most common nuclides (Flynn et al., 1964; Horrocks, 1964; McDowell, 1971).

Alpha counting and spectroscopy by liquid scintillation has some obvious advantages over other methods. Since the alpha-emitting nuclide is homogeneously dispersed in the scintillator medium, the problems of sample self-absorption are avoided, allowing 100% counting efficiency and facilitating sample preparation. All the difficulties associated with plate preparation and detector geometry are eliminated, resulting in better reproducibility from sample to sample (of the same type) by liquid scintillation than can routinely be obtained by either electroplating or drying a sample on a plate. The possibility of obtaining alpha energy spectra from a liquid scintillator containing a dissolved or dispersed alpha emitter (Horrocks, 1964; McDowell, 1971; Horrocks and Studier, 1964) is less well recognized. Such spectra can be obtained when an aqueous solution is incorporated in an organic scintillator with an emulsifying agent. However, there appears to be considerable advantage in energy resolution if the nuclide is incorporated into the scintillator by the use of an organic-soluble complexing agent (Flynn et al., 1964; Horrocks, 1964; Horrocks, 1974b), and even better energy resolution results from the use of an all-organic scintillator containing no aqueous-phase-accepting components (Horrocks, 1964; McDowell, 1971; Horrocks and Studier, 1964; Ihle et al., 1965).

In spite of the above advantages, alpha counting by liquid scintillation has failed to gain wide acceptance, partly because alternative methods such as gas-flow proportional counting, zinc sulfide scintillators, and surface barrier detectors have become established as the conventional

methods and partly because of some disadvantages of liquid scintillation as it is ordinarily used and perceived. These disadvantages are primarily related to two areas: (1) determination of background, and (2) energy resolution and identification.

Beta or gamma radiation produces more light for the same amount of energy absorbed in a liquid scintillator than does alpha radiation, resulting in a serious overlap of beta-gamma and alpha spectra. Figure 1 illustrates the type of energy (pulse-height) distribution observed for alpha, beta, and gamma emissions. The beta and gamma radiation may arise from the alpha nuclide decay scheme, may be introduced to the scintillator as an impurity along with the sample, or may be of external origin (including the sample container and phototube face). Therefore, the usual method of background counting and subtraction will frequently be inadequate in liquid scintillation counting.

Energy resolution constraints arise from a variety of sources. Beta liquid scintillation equipment usually has limited energy resolution capability for alpha particles because highly refined energy resolution is not needed for the broad zero-to-maximum energy distribution of beta spectra. Further, quenching effects tend to complicate the calibration of an alpha energy scale in liquid scintillation. When a

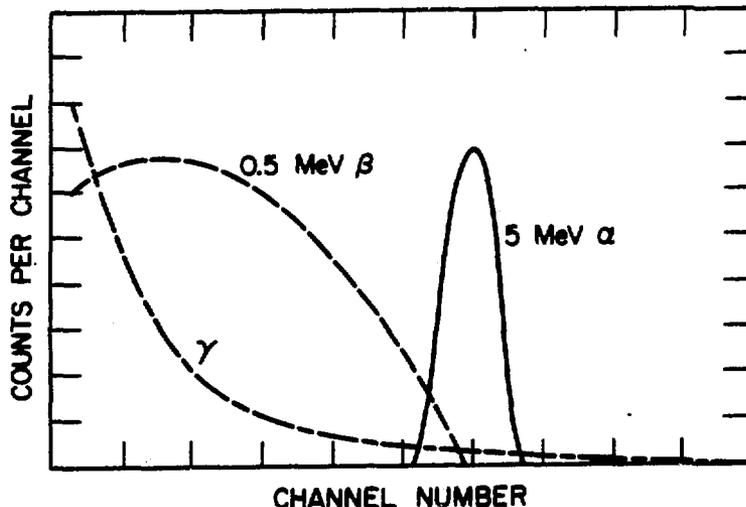


FIGURE 1. Typical liquid scintillation spectra of alpha, beta, and gamma radiation.

sample for alpha counting is prepared in the manner normally used for beta counting, the addition of water, acids, salts, or other impurities changes the scintillator response and shifts the alpha spectrum up or down scale; thus, although the alpha counting efficiency usually remains at 100%, the ability to identify alpha energies is impaired or lost.

II. RECENT ADVANCES

Increased interest in the use of liquid scintillation for alpha counting became evident beginning in the early 1960s. During the next 10 to 15 years, a number of approaches were made to solving the problems associated with alpha counting and spectrometry.

A. *Modification in Beta Liquid Scintillation Systems*

Commercial instrumentation and aqueous-phase-accepting scintillators designed for beta counting were used in much of the early reported work, and corrections for beta-gamma interference were made by simple subtraction or graphical subtraction, often aided by a foreknowledge of the interfering nuclides(s) present (Horrocks, 1966; Bogen and Welford, 1971; Lindenbaum and Judd, 1969). Such procedures are often quite useful for laboratory-generated (or other) samples in cases where it is known that a single alpha emitter of relatively high count rate is present and background interferences are known or negligible. The automatic sample-changing equipment usually included in commercial beta liquid scintillation systems is also often an advantage. In some instances, commercially available equipment was modified to yield better energy resolution (Hiebert et al., 1977) or sample preparation methods or scintillators were changed to reduce background or improve energy resolution (Ihle et al., 1965; Toribara et al., 1977; Gorbushina et al., 1973; Ghysels, 1972; Keough and Powers, 1970; Darall et al., 1973) in such equipment. In at least two cases, commercial instrumentation was modified to allow electronic discrimination between alpha pulses and beta or gamma pulses (Hiebert et al., 1977; Buchlela et al., 1974), allowing the more slowly decaying (longer) pulses produced by alpha particles in a liquid scintillator to be accepted and the beta- and gamma-produced pulses to be rejected.

B. All-Organic and Extractive Scintillators

A few investigators recognized the advantages of introducing the nuclide into the scintillator by first extracting it into an organic phase containing an organic-soluble complexant or a solvent extraction agent and combining a portion of the organic extract with the scintillator (Horrocks, 1964; Horrocks and Studier, 1964, Ihle *et al.*, 1965; Horrocks, 1966). Such a procedure reduces quenching effects and allows the response of the scintillator medium to be more constant from sample to sample. A further development along these lines involved including such a reagent in the scintillator solution itself so that the nuclide can be extracted directly into it from an appropriate aqueous phase (McDowell, 1971; Keough and Powers, 1970; Kim and MacInnis, 1971; Ham *et al.*, 1977). This mixture is called an "extractive scintillator." In addition to the advantage of simplicity, this procedure achieves nearly complete constancy of the scintillating medium. Aqueous solutions and all agents for incorporating them into the scintillator (e.g., solubilizing, dispersing, gelling agents) are avoided, eliminating their large and variable quench effects. The addition of increments of organic extractant solution is also avoided. In effect, only the trace nuclide is added to the standardized scintillator; consequently, the pulse height and corresponding peak position for a given alpha energy can be made constant.

At Oak Ridge National Laboratory (ORNL), the concept of the extractive scintillator has been used extensively (McDowell and Henley, 1972; McDowell, 1975, McDowell *et al.*, 1974; Perdue *et al.*, 1976; McDowell and Weiss, 1977; Bower *et al.*, 1978; McDowell and Coleman, 1974; Bower *et al.*, 1979). A number of extractants have been tested and their quenching tendencies evaluated (McDowell and Coleman, 1973). A few typical extractants with their properties are listed in Table I. An extractive scintillator composition of 0.1 to 0.2 moles/l of the extractant, 150 to 200 g/l of naphthalene, and 4 to 5 g/l of a single fluor was found to be most suitable for both pulse-height and pulse-shape resolution properties. Of the various fluors tested, ONPO [2-(1-naphthyl)-5-phenyl-oxazole] and PBBO [2-(4'-biphenyl)-6-phenylbenzoxazole] were found to have the best pulse-height response and energy resolution properties (McDowell *et al.*, 1974). The extractant used in the scintillator (and the aqueous medium from which the extraction is made) can often be chosen so as to provide a degree of selectivity for the nuclide of interest; thus the

TABLE I. Extractants for Use with Extractive Scintillators

Extractant	Alpha emitter extracted	Quenching
Di(2-ethylhexyl) phosphoric acid	All cations; trivalent actinides, thorium, and uranyl best	Very slight
Tertiary amine sulfate (trioctyl, triisooctyl, or triisodecyl)	Uranium(VI)	Very slight
Primary amine sulfate (branched 16- to 20-carbon compound)	Uranium(VI), thorium	Very slight
Tertiary amine nitrate	Plutonium(IV)	Moderate to severe
Tertiary amine chloride	Polonium, uranium(VI)	Moderate to severe ^a
Trioctylphosphine oxide	Uranium(VI), thorium, plutonium(IV)	Depends on anion present; ^a slight for sulfate, moderate to severe for nitrate or chloride
Tributyl phosphate	Uranium(VI), thorium, plutonium(IV)	

^a May be necessary to strip and reextract into scintillator containing di(2-ethylhexyl)-phosphoric acid to obtain good energy and pulse-shape resolution.

extraction into the scintillator becomes the last step in a separation scheme that aids in identification of the nuclide (McDowell, 1975; McDowell et al., 1974; McDowell and Weiss, 1977; Bouwer et al., 1979).

C. Detectors and Electronics

Concurrent with the improvement in scintillators, a number of improvements in detectors and electronics were made. The combination of these developments allowed marked advances in energy and pulse-shape resolution in alpha liquid scintillation spectrometry. It was recognized early that a single phototube was sufficient for alpha counting (Horrocks, 1964; McDowell, 1971; Horrocks and Studier, 1964; McKlveen and Johnson, 1975). The dual phototube-coincidence arrangement used to reject random noise pulses and lower the energy detection threshold for soft betas is not needed, nor is the refrigeration chamber for the detector that is used for the same purpose.

The optical requirements for good alpha energy resolution appear to be as follows: (1) A scintillation event must appear the same to the phototube irrespective of where it occurs in the sample. (2) Since the photocathode is not homogeneous, but varies in sensitivity from place to place, the light from any scintillation anywhere in the sample must be spread evenly over the face of the phototube. (3) Light collection must be efficient since the resolution is a statistical function of the quantity of light collected.

Horrocks and Studier (1964) were among the first to seek optimization of sample size, light coupling arrangements, and reflectors to increase energy resolution. Although these innovations were made to lower the counter background and improve efficiency for soft beta emissions from ^{241}Pu , the presence of alpha-emitting plutonium isotopes in the same sample allowed the investigators to recognize possibilities for improved alpha energy resolution. Subsequent work confirmed that small samples, good optical coupling, and efficient reflectors were necessary to obtain good alpha energy resolution. Although later work by others showed some progress in optimizing sample size and light-collection arrangements (Horrocks, 1964; McDowell, 1971; McKlveen and Johnson, 1975; McKlveen and McDowell, 1976), it is not apparent that the best conditions have been attained. However, Hanschke (1972), who has made an extensive study of the effects of reflector shape, sample volume and shape, and photocathode variability, concludes that a small-volume sample and a reflector which is a section of a sphere give the best resolution for a given phototube situation. At ORNL good

light-collection and good pulse-height resolution have been obtained by using a 1.0- to 1.5-ml sample contained in a standard 10 x 75 culture tube and held between the face of the phototube and flat-white reflector shaped as a section of the inside of a sphere (Figure 2). The space between the reflector and phototube face is filled with silicone oil to minimize reflecting surfaces at refractive index discontinuities. A large number of sample size and reflector arrangements were evaluated before arriving at the configuration shown in Figure 2. Some of this work has been reported previously (McKlveen and McDowell, 1976).

General improvements in multiplier phototubes have contributed significantly to the ability to develop an alpha liquid scintillation detector with desired energy resolution characteristics. Only limited tests of available phototubes for this application have been made (Thorngate and Christian, 1977). At ORNL, two flat-faced, 2-in., bialkali phototubes have been found useful: the RCA 4523 and the EMI 9840A. Other tests indicate that the RCA 8575 phototube has favorable characteristics; however, differences between phototubes of the same type are often greater than those observed for different types.

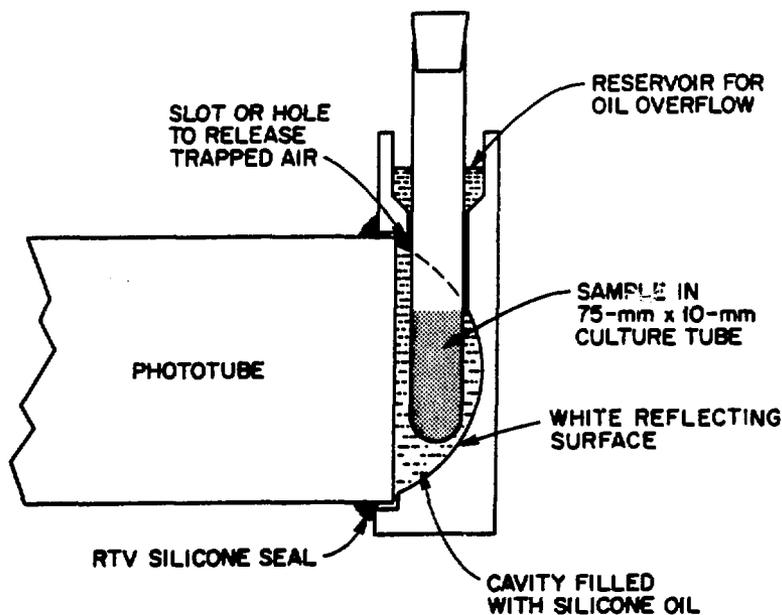


FIGURE 2. Cross section of the basic design for the high-resolution liquid scintillation detector.

Electronics systems for processing the signal generated by a phototube used in alpha liquid scintillation work have varied from the standard circuits supplied with commercial beta liquid scintillation equipment, through various modifications of such equipment, to completely constructed systems designed for alpha spectrometry and pulse-shape discrimination. Linear amplifiers and preamplifiers of the type generally used for gamma spectrometry with sodium iodide crystal--phototube combinations appear to be quite suitable for alpha liquid scintillation spectrometry. Such equipment has been used by several investigators (Horrocks, 1964; McDowell, 1971; Horrocks and Studier, 1964; Horrocks, 1966; McKlveen and Johnson, 1975; Seliger, 1960). Typically, a scintillation preamplifier is followed by a linear amplifier that feeds the signal into a multichannel analyzer. Alpha energy resolution typical of such electronics in conjunction with a single bialkali phototube with reflector and small-volume sample as described previously is shown in Figure 3. A spectrum of the

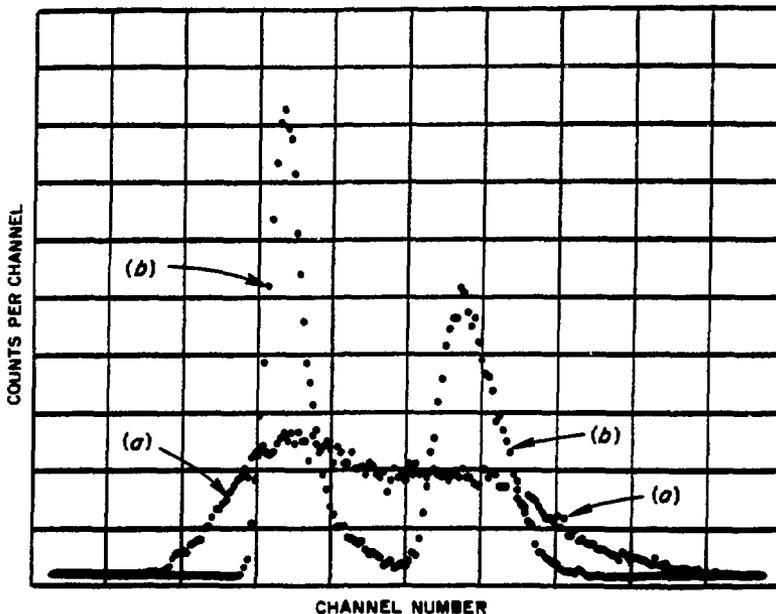


FIGURE 3. Comparison of spectra from (a) a commercial beta liquid scintillation spectrometer and (b) from high-resolution alpha liquid scintillation spectrometer. The same pair of alpha emitters (^{232}Th ; 4.01 MeV and ^{239}Pu ; 5.15 MeV) were used in the same scintillator in both cases.

same pair of nuclides in the same scintillator recorded from the output of a commercial beta liquid scintillation spectrometer is included for comparison. The higher energy resolution for the alpha scintillation counting is due partly to the more efficient light-collecting arrangement and partly to the improved electronics system.

D. Pulse-Shape Discrimination

One of the most important developments in electronics for alpha liquid scintillation counting is the use of electronic pulse-shape discrimination to separate and reject pulses produced by beta and gamma radiation. The techniques of pulse-shape discrimination are well known and have been extensively applied to the rejection of gamma background in neutron spectrometry (Horrocks, 1970; Winyard et al., 1971) and to the isolation of fission events in liquid scintillation counting (Horrocks, 1963). Only limited application has been made, however, to the separation of beta- and gamma-produced pulses from alpha-produced pulses arising from a liquid scintillator (Lindebaum and Judd, 1969; Darall et al., 1973; McKlveen and Johnson, 1975; Thorngate et al., 1974).

Pulse separation in pulse-shape discrimination is achieved by taking advantage of the slower decay of the light pulse produced by alpha particles compared with that produced by beta or gamma radiation. Thus, the effectiveness of pulse-shape discrimination, like that of pulse-height resolution, depends upon several factors. For example, the scintillator must be of high efficiency and free of chemical quenching materials, particularly dissolved oxygen. In addition, the optical coupling between sample and phototube must be optimum. While reflectors yielding good pulse-shape resolution apparently always give good pulse-height resolution, the reverse is not necessarily true (McKlveen and McDowell, 1976). Finally, the phototube and amplifiers must be able to retain in their output signal the time difference in light pulse produced in the scintillator by the different types of radiation.

Several types of pulse-shape discrimination electronic circuits have been described in the literature (McBeth et al., 1971). A very efficient and reliable arrangement used at ORNL is that developed by Thorngate (Thorngate et al., 1974). A block diagram of the commercial components comprising this system, illustrating pulse forms observed at different locations, may be seen in Figure 4. A very simple two-unit-wide pulse-shape analyzer, designed primarily for alpha spectrometry, has also been built (Thorngate, 1978).

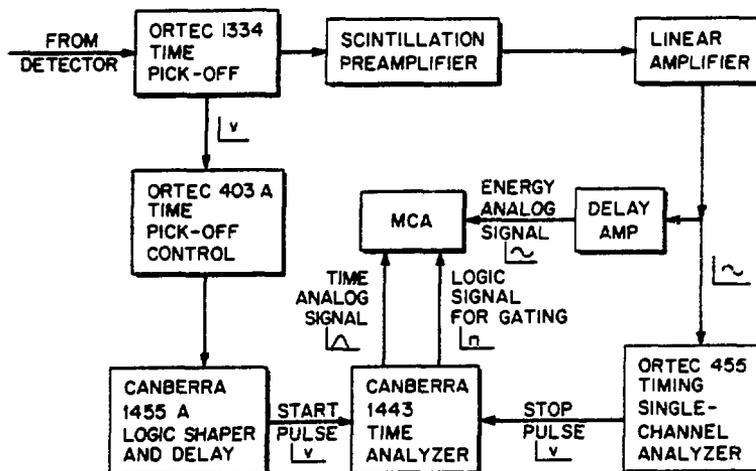


FIGURE 4. Block diagram of electronics for pulse-shape discrimination.

Figure 5 shows the results of using pulse-shape discrimination circuitry to resolve the mixed beta, gamma, and alpha pulses from a sample containing ^{232}Th and its daughters (spectrum A). Spectrum B is a time spectrum showing the beta-gamma pulses (left) separated from the alpha pulses. When the alpha pulses are selected by a single-channel analyzer and a logic pulse derived from them used to gate the multichannel analyzer, spectrum C, devoid of the beta-gamma continuum, is obtained. Backgrounds of 0.01 cpm are easily obtained with pulse-shape discrimination and with careful work it has been possible to achieve backgrounds as low as 0.002 cpm under a typical alpha peak. With samples containing internal beta and gamma radiation, it is possible to remove 99% or more of the beta-gamma component and retain a similar percent of the alpha counts.

III. APPLICATIONS

The degree of complexity and sophistication necessary in alpha liquid scintillation is dictated by the properties of the sample (physical and chemical forms, impurities present, type of nuclide to be counted,) and the accuracy of the results required. Several of the possible options are listed in Table II.

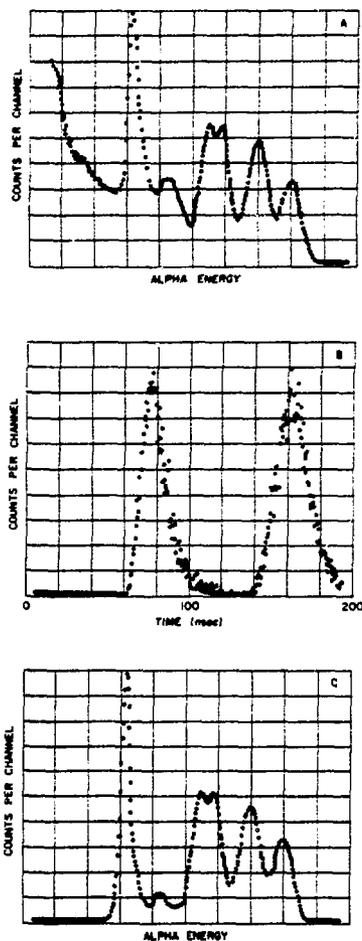


FIGURE 5. Spectrum of ^{232}Th and daughters (a) including beta-gamma component and (b) using pulse-shape discrimination to remove beta and gamma pulses.

TABLE II. Options for Alpha Liquid Scintillation (α LS)

Method	Capability	Advantages	Disadvantages
<ul style="list-style-type: none"> • Aqueous sample • All-purpose scintillator • β LS counter 	<ul style="list-style-type: none"> • Gross α count of relatively high count rate 	<ul style="list-style-type: none"> • Easy sample prep. • Available equip. and scintillator 	<ul style="list-style-type: none"> • Little energy resolution • Variable quench • Variable background
<ul style="list-style-type: none"> • Extractive scintillator • β LS counter 	<ul style="list-style-type: none"> • Gross α count of somewhat lower count rate 	<ul style="list-style-type: none"> • Less background var. • Reproducible quenching 	<ul style="list-style-type: none"> • Little energy resolution • Still relatively high background
<ul style="list-style-type: none"> • Extractive scintillator • α LS spectrometer 	<ul style="list-style-type: none"> • α spectra 200- to 300-keV FWHM • Lower detn. limit 	<ul style="list-style-type: none"> • Lower external bkgd. • Usable energy spectrum 	<ul style="list-style-type: none"> • More sample prep. • β-γ from nuclide or daughters visible
<ul style="list-style-type: none"> • Extractive scintillator • α LS spectrometer • Pulse-shape disc 	<ul style="list-style-type: none"> • α spectra • Low detn. limit 	<ul style="list-style-type: none"> • Internal β-γ rejected • Low, reproducible bkgd. 	<ul style="list-style-type: none"> • Additional sample prep. • Additional electronics

If the samples contain only one alpha-emitting nuclide and minimum, or known amounts, of beta and gamma emitters and the count rate is sufficiently above background, commercial beta liquid scintillation equipment and scintillators are an appropriate and convenient means of alpha counting. Useful background reduction and scintillator-response standardization can be achieved by the use of an extractive scintillator in conjunction with a beta liquid scintillation counter.

The ability to isolate alpha-emitting nuclides chemically and identify them by a combination of chemical selectivity and alpha spectrometry may be realized by using a combination of standard chemical separations, an extractive scintillator, and high-resolution alpha liquid scintillation spectrometry. For example, plutonium can be extracted selectively from nitric acid digests of a variety of materials and then stripped and reextracted into a scintillator (1.2 to 1.5 ml) for subsequent counting in a high-resolution detector (McDowell et al., 1974). Uranium and thorium can be extracted together, stripped into an aqueous phase, and reextracted selectively into separate scintillators (Bouwer et al., 1979). The trivalent actinides can be separated by conventional methods and then extracted into a scintillator containing di(2-ethylhexyl)phosphoric acid (McDowell, 1971).

The lower limit of detection and the accuracy of counting by these methods are, as in other counting methods, determined by both the sample count rate and the background count rate (Horrocks, 1974c). Applicable background count rates are decreased by better energy resolution and by pulse-shape discrimination; thus, the ability to count low-level samples improves in going from a beta liquid scintillation counter with an all-purpose, aqueous-accepting scintillator to a high-resolution alpha liquid scintillation spectrometer with extractive scintillator and pulse-shape discrimination (see Table II). In the latter case, for example, a background count rate and sample count rate of 0.01 cpm would yield a determination with a standard deviation of 55% in 1000-min counting time, while a sample count rate of 10 cpm with a background of 0.01 cpm would yield a standard deviation of 1.0%.

IV. FUTURE POSSIBILITIES AND PROBLEMS

There is a great need for a simple, rapid, and accurate method of alpha counting and spectrometry. Unfortunately, the nature of alpha radiation makes the development of such a method difficult. However, liquid scintillation methods

with some modification in sample preparation, scintillator, and counting equipment may be the most practical approach. Alpha-emitting nuclides must still be chemically separated from any large amounts of interfering material, but preparation of a liquid scintillation sample usually demands much less time and fewer steps than does preparation of a plate for surface barrier work. Nevertheless, preparation of a large number of environmental survey samples, for example, will require considerable effort. At ORNL, several semi-automated methods have been explored for reducing the labor involved in sample preparation. Standard methods of digesting or evaporating several samples simultaneously under infrared lights and fuming with acid using thermostatically controlled heating equipment are used to eliminate the necessity of constant attention by an analyst. Multiple solvent extraction equilibrations may be performed by clamping the separatory funnels on a rod that can be rotated at about 20 rpm.

A small separatory funnel or other vessel is required if extraction into the small volume of scintillator for high-resolution work is to be done. Several devices are being tested that may allow the extractive scintillator to be equilibrated with the sample under an oxygen-free atmosphere followed by counting in the same disposable vessel. The equilibrated scintillator (separated from the aqueous phase) would then be ready for use with pulse-shape discrimination.

The lack of commercial availability of the detector, and of some of the extractants listed in Table I in sufficient purity for good liquid scintillation work, constitutes an obstacle to widespread use of the more-refined forms of liquid scintillation alpha spectroscopy in service analytical laboratories. A number of laboratories are presently using such procedures and equipment; however, it is hoped that sufficient interest will produce commercial supplies as was the case in the early use of beta liquid scintillation. Adequate descriptions of purification methods for the quantities of reagents needed for experimental or research work are given in references in this paper (McDowell and Henley, 1972; McDowell, 1975; McDowell et al., 1974; Perdue et al., 1976; McDowell and Weiss, 1977; Bouwer et al., 1978; McDowell and Coleman, 1974; Bouwer et al., 1979; McDowell and Coleman, 1973).

It is not clear whether the practical limit of energy resolution for alpha liquid scintillation work has been reached. The relevant factors are associated either with the scintillator or with the detector since electronic signal processing is clearly not a limit. Considerable improvements have been made in resolution by choosing the purest and most

effective ingredients (McDowell, 1975; McDowell et al., 1974; McDowell and Coleman, 1973), but it appears difficult to extend the capability beyond 200- to 300-keV FWHM. This resolution, which corresponds to 4 to 6%, is somewhat better than can be obtained in gamma spectrometry by optimizing a phototube--sodium iodide crystal combination.

The overall efficiency of converting ionizing radiation energy to electrical pulses is probably the dominating factor in determining resolution. Thus, any improvements in scintillator, reflector, and phototube efficiency would be expected to improve resolution. A phototube designed specifically for alpha liquid scintillation spectrometry might offer considerable advantages. Present phototubes are designed to react to light from distant or facing light sources rather than light from a curved solid volume. A phototube with photocathode and dynode elements arranged to surround the sample might respond more effectively and reproducibly to light flashes from a small volume of liquid scintillator.

Most detectors presently in use for alpha liquid scintillation spectrometry do not incorporate sample changes, partly because of their experimental nature and partly because they are not really needed for low-count-rate samples. Sample changers using the detector and sample configuration described for high-resolution alpha liquid scintillation spectrometry have been constructed; however (Thorngate, 1977), like the detector, they are not commercially available.

V. CONCLUSIONS

The concept of liquid scintillation counting offers inherent advantages for alpha counting and spectrometry with regard to sample preparation and counting efficiency. Recent improvements in sample preparation, scintillators, and equipment designed to optimize the system for alpha assay indicate that rapid, accurate, and efficient methods of alpha counting and spectrometry are possible with liquid scintillation. Further improvements are needed in sample preparation methods, scintillator efficiency, and detector (primarily phototube) design to allow widespread use of alpha liquid scintillation spectrometry.

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