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LIQUID SCINTILLATION ALPHA COUNTING AND SPECTROMETRY AND ITS APPLICATION TO BONE AND TISSUE SAMPLES*

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ABSTRACT

Three methods for determination of alpha-emitting nuclides using liquid scintillation counting are compared, and the pertinent literature is reviewed. Data showing the application of each method to the measurement of plutonium concentration in tissue and bone samples are presented. Counting with a commercial beta-liquid scintillation counter and an aqueous-phase-accepting scintillator is shown to be accurate only in cases where the alpha activity is high (several hundred counts/min or more), only gross alpha counting is desired, and beta-gamma emitters are known to be absent from the sample or present at low levels compared with the alpha activity. Counting with the same equipment and an aqueous immiscible scintillator containing an extractant for the nuclide of interest (extractive scintillator) is shown to allow better control of alpha peak shift due to quenching, a significant reduction of beta-gamma interference, and, usually, a low background. The desirability of using a multichannel pulse-height analyzer in the above two counting methods is stressed. The use of equipment and procedures designed for alpha liquid scintillation counting is shown to allow alpha spectrometry with an energy resolution capability of 200 to 300 keV full-peak-width-at-half-peak-height and a background of 0.3 to 1.0 counts/min/ℓ, or as low as 0.01 counts/min if pulse-shape discrimination methods are used. Methods for preparing animal bone and tissue samples for assay are described.

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INTRODUCTION

Although the principal use of liquid scintillation counting is the assay of low-energy beta emitting nuclides, it has been known for more than two decades that alpha particles could be counted effectively by this technique (1). The use of liquid scintillation for alpha counting is attractive because of its 100% counting efficiency and simplicity of sample preparation; and, in the late 1950's and early 1960's several workers have made applications of the method (2-6). Widespread use of alpha liquid scintillation counting has been hindered because of the high background count and very poor energy resolution for alpha energies associated with the usual beta liquid scintillation methods.

In 1964 and 1965 Horrocks (6) in this country and Ihle et al. (7) in Germany demonstrated that a useful degree of alpha energy resolution could be attained by liquid scintillation methods. However, little practical use was made of these high-resolution methods until recently because the detectors and associated electronics needed are not commercially available and because the nuclide must be placed in the scintillator as an organic-soluble complex. This latter requirement has, however, led to the use of liquid-liquid extraction as a convenient means for both isolating the desired nuclide and placing it in a water-immiscible scintillator solution. Several workers have developed liquid scintillation counting procedures for alpha-emitting nuclides based on liquid-liquid extraction separations and/or the use of a water-immiscible scintillator containing an extractant (extractive scintillator), both for use with beta liquid scintillation counters (2,6-10), and for the application of high resolution alpha liquid scintillation spectrometry (11, 12). Even more recently, the introduction of pulse-shape discrimination electronics has allowed the separation of alpha pulses from beta- or gamma-produced pulses in liquid scintillation systems and, hence, has made dramatic reduction in background count in alpha counting possible (13, 14). Figure 1 shows a direct comparison between the usual beta liquid scintillation counter and the high-resolution detector as regards energy resolution and the separation of alpha pulses from beta-gamma pulses.

This paper will discuss and compare the various forms of liquid scintillation for alpha counting and their advantages and disadvantages. Because of the increasing importance of studies of animal metabolism of alpha-emitting nuclides, particularly plutonium, and because of some studies of plutonium uptake currently in progress in our laboratories,

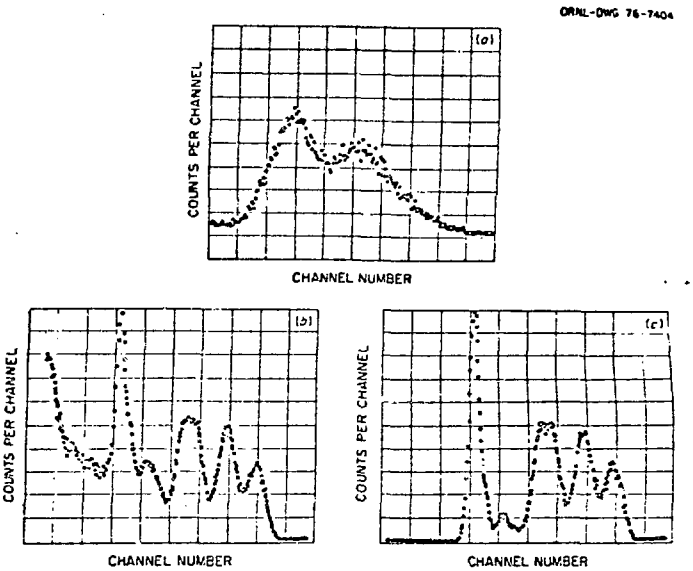

Fig. 1. Spectrum of ^{232}Th and daughters, ^{228}Th, ^{224}Ra, ^{220}Rn and ^{216}Po with some ^{230}Th as seen (a) by a commercial β liquid scintillation counter and (b) by the high-resolution detector without β-γ rejection by pulse-shape discrimination and (c) with pulse-shape discrimination.

liquid scintillation counting of plutonium in animal tissue and bone samples will be emphasized.

EXPERIMENTAL

Equipment

The commercial beta-liquid-scintillation detector used was a Packard Model 3214. An interface to a multichannel analyzer was arranged according to a diagram provided by D. C. Bogen (15) of the Health and Safety Laboratory, New York. Any of several commercial beta liquid scintillation counters could be used; and some types presently available have provisions for interfacing to a multichannel analyzer. It is desirable that the pulse-height information for the multichannel analyzer be taken from the scintillation counter circuitry at some point following the upper and lower discriminator and that it be linear with light output. This permits direct observation of the effect of adjusting the

discriminators via the multichannel analyzer display and simplifies optimization of the settings to include all of the alpha peak while rejecting as much background as possible.

The high-resolution liquid scintillation alpha spectrometer was constructed from a detector consisting of an RCA 4523 or an EMI 9840A phototube sealed to a reflector-sample holder unit, as shown in Figure 2. Standard preamplifiers and linear amplifiers were used to present the pulse-height information provided by the phototube to a multichannel analyzer. Pulse-shape discrimination circuitry was assembled from standard components. More detailed descriptions of the high-resolution detector and associated electronics and of the pulse shape discrimination equipment are given in earlier publications (16, 17).

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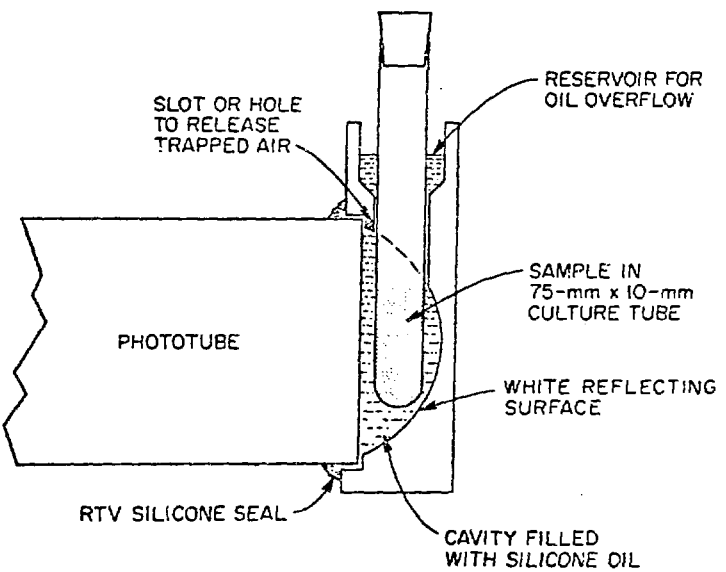

Fig. 2. Detector assembly for high-resolution alpha liquid scintillation spectrometer.

Reagents

The extractant used in the scintillator, di(2-ethylhexyl)-phosphoric acid (HDEHP), was obtained from the Virginia Carolina Chemical Company. As received, this reagent contained about 1%

impurities and was slightly colored. It was purified by a procedure involving precipitation of the copper salt of HDEHP. Details of this procedure are available in previous publications (18, 19). The tertiary amine, Adogen 364, was obtained from the Ashland Chemicals Company and was purified by single-stage high vacuum distillation. Naphthalene used in the scintillator was purified by resublimation or, for some tests, by zone refining. The scintillator, PBBO [2-(4'-biphenylyl-6-phenylbenzoxazole] was obtained from Eastman Chemical Products, Incorporated, and used without further purification.

Plutonium-239 was obtained from the Isotope Division, ORNL. All other chemicals were of the usual reagent grade.

DESCRIPTION AND DISCUSSION OF PROCEDURES

Sample preparation

Small bone and tissue samples (< 25 g) are placed in solution with concentrated nitric acid and small amounts of 30% hydrogen peroxide. Gentle heat, to the entire container so as to prevent bumping, is provided primarily from radiant heaters. Digestion is continued until a clear solution is obtained. Samples with only small amounts of residual salts should not be allowed to go dry since plutonium and other nuclides may sorb on glass surfaces or otherwise become difficult or impossible to redissolve.

Larger-sized samples (> 25 g) are dissolved by successive treatments with concentrated nitric acid and 30% hydrogen peroxide, each treatment being followed by evaporation to dryness and heating to 450°C overnight in a furnace. If the residue is not white, the treatment with concentrated nitric acid and 30% hydrogen peroxide is repeated, followed by again heating overnight in the furnace. This series of steps usually produces a white ash, free of carbon and organic material. The ash is then dissolved in sufficient 2 M HNO_3, and the sample is subsequently treated by whatever method (outlined below) seems best according to the level of activity anticipated in the sample. Heating to 450°C does not cause a loss of plutonium in the larger-sized tissue samples, probably because of the presence of sufficient amounts of salts so that the plutonium enters the mineral lattice rather than being adsorbed on the surface of the glass.

After the sample has been completely dissolved, the resulting clear nitric acid solution is treated in one of three ways, depending on the counting method to be employed: (a) If the sample is known to contain high levels of activity and is to be assayed using an aqueous-phase-

accepting scintillator, the solution is diluted to a known volume after digestion and an aliquot is taken for addition to the scintillator. (b) If the sample is to be treated by an anion exchange separation (liquid or resin, see below), the solution is diluted or treated in such a manner as to yield the desired nitric or hydrochloric acid concentration. (c) If the sample is intended for direct extraction into a scintillator containing HDEHP, sufficient perchloric acid is added to the solution to give a final solution that will be 0.1 to 0.2 N in perchloric acid after all the metal ions present in the sample (from whatever source) have been converted to perchlorate salts. The nitric acid is evaporated with slightly increased heat (150 to 170°C), leaving only perchlorates and perchloric acid. This procedure ensures complete destruction of all remaining organic material and allows volume reduction to a salt solution containing a controlled amount of acid without the risk of the sample ever becoming dry. In this procedure, care should be taken to ensure that only very small amounts of organic material, if any, remain when the perchloric acid is added and that nitric acid is present when the perchloric acid is added. If these precautions are observed, there is no danger of an explosive reaction.

To facilitate plutonium extraction into HDEHP, 2 ml of saturated aluminum nitrate per gram of sample and sufficient water to make 5 ml per gram of sample is added to the sample while it is still hot.* A 5- to 8-ml volume of such a sample can be added directly to a 20-ml vial of the kind commonly used for beta liquid scintillation counting and extracted directly into 10 ml of an extractive scintillator in the vial (see below), or larger extractions can be made using a separatory funnel. However, phase ratios of ~ 1:1 organic: aqueous should be maintained to obtain quantitative plutonium recovery. Extractions from this medium for high-resolution alpha liquid scintillation counting or for pulse shape discrimination work are, however, not usually successful because of quenching due to extracted nitrate ion (nitric acid) and from extraction of unwanted metal ions such as iron and calcium. Separations designed to prepare samples for the high-resolution, low background detector will be described in the section dealing with that method of counting.

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*Aluminum nitrate improves plutonium extraction by, (a) nitrate stabilization of Pu(IV), (b) complexing phosphate, and (c) increasing ionic strength.

Counting in a beta liquid scintillation counter using an all-purpose scintillator

Using an aqueous-phase-accepting scintillator (e.g., one composed of 160 g of naphthalene, 10 g pf PPO (2,5-diphenyloxazole), 0.1 g of POPOP (2,2-p-phenylene-bis-5-phenyloxazole), 385 ml xylene, 385 ml dioxane, 230 ml ethyl alcohol and 1000 ml of Triton x-100), up to 1 ml of a highly salted aqueous phase such as that resulting fron nitric acid dissolution of a tissue or bone sample, with or without the added perchloric acid, can be incorporated directly into the scintillator. Such a sample can be counted in a commercial beta liquid scintillation counter. For gross alpha counting of samples with a sufficiently high count rate (e.g., at least several hundred counts/min), this counting method is adequate if measures are taken to ensure that the alpha peak is within the pulse-height range observed. Varying amounts of salt, acid, or even water cause the pulse-height response of the scintillator to change, thus shifting the position at which the alpha pulses appear. The peak position may be found by counting through a narrow window as the available pulse-height range is scanned, but the use of a multichannel analyzer connected to the counter is more convenient and also allows visual differentiation between alpha and beta-gamma spectra. Unless very similar amounts of salt, water, and acid are present in each sample, the position of the alpha peak should be monitored for each sample to ensure reproducible counting efficiency.

Backgrounds of 20 to 30 counts/min plus a contribution of 10 to 20 counts/min from ^{40}K in each gram of tissue are usually encountered in alpha counting in this way. Some reduction of background count can be effected by careful adjustment of the upper and lower discriminators to include the alpha peak and reject as much of the beta-gamma contribution as possible; however, this advantage is gained at the cost of a rather time-consuming individual discriminator adjustment for each sample because of variable quenching.

The accuracy with which a sample can be counted by this method is, of course, determined by the usual statistical relationship that includes the background count; therefore, the counting accuracy decreases rapidly as the net sample count decreases toward, and falls below, the background count. A total count equal to at least twice the background count is usually considered the practical lower limit for counting with reasonable accuracy. With samples prepared as above, it must be remembered that ^{40}K and any other beta- or gamma-emitting nuclides in the sample must be considered as background and in these cases background must be determined

individually for each sample from a spectrum of that sample or it must be assumed that all the samples are alike in this respect.

No energy resolution or pulse-shape discrimination is possible with this counting method.

Counting in a beta-liquid-scintillation counter using an extractive scintillator

A scintillator containing 161 g of HDEHP, 80 g of naphthalene, and 4 g of PBBO (or 5 g of PPO) per liter of toluene can be used to extract the plutonium (or most other alpha-emitting nuclides) from a properly prepared sample. Figure 4 shows the percent plutonium recovered from a 5-ml sample containing 1 g of dissolved bone as a function of HDEHP concentration. Recovery from tissue samples is easier than from bone and usually can be done with lower HDEHP concentrations. This approach to alpha-liquid scintillation counting, which has been used by several workers (8-10, 12) has important advantages: (1) The background contribution from ^{40}K in the sample is reduced dramatically since HDEHP does not extract potassium and the interaction of the ^{40}K beta with the scintillator is greatly reduced. (2) The scintillator is more reproducible and the pulse-height response to a given alpha is more nearly the same, thereby reducing the need for window adjustment for each sample. (3) Finally, and perhaps most importantly, an extracted nuclide can be stripped (see in following section) and reextracted into the high-resolution

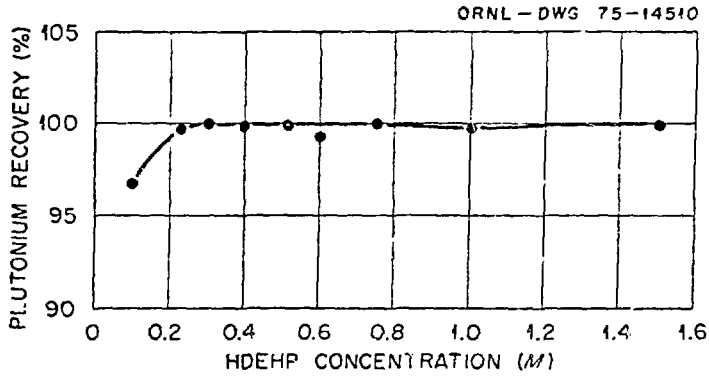

Fig. 3. Percentage recovery of plutonium from 5 ml samples containing 1 g of bone in perchlorate medium as a function of HDEHP concentration in the scintillator.

HDEHP concentration in the scintillator.

scintillator for analysis in the high-resolution detector, where energy resolution and much lower backgrounds can be attained.

The sample is prepared for extraction as outlined under "Sample preparation" [method 3] and transferred to a standard 20-ml scintillator vial; then 10 ml of the extractive scintillator is added. The vial is shaken for 1 to 2 minutes, the phases allowed to separate, and the vial placed in the counter. Counting can be done without separation of the aqueous phase. It has been shown by Keough and Powers (8), and Horrocks (11), that interphase counting efficiency varies from 2% for energetic betas to less than 0.1% for alpha, gamma and low-energy beta emitters remaining unextracted in the aqueous phase. Although there is some quenching from extracted nitrate, there will be only minor variation in the pulse-height response of scintillator-samples prepared and extracted in this way. Although the pulse-height response is more consistent than with an aqueous-phase-accepting scintillator, it is still desirable to use a multichannel analyzer to monitor the position of the alpha peak.

Of twelve 400 to 800 count/min samples run as described above, the average plutonium recovery was 100.5% with a standard deviation of 1.2%.

Under the best conditions some alpha energy resolution is possible using this procedure. The full-peak-width-at-half-maximum-peak height (FWHM) is usually 0.9 to 1.0 MeV. Thus, it is sometimes possible to distinguish, and count separately, alpha energies that differ by more than 1 MeV.

Separation of alpha from beta-gamma pulses is usually not possible by this procedure either on an energy basis or via pulse-shape discrimination (20). Beta or gamma interactions with the scintillator produce light approximately 10 times more efficiently than do alpha particle interactions. This difference in light-producing efficiency plus the fact that both beta and gamma radiation produce a continuous pulse height distribution from zero to some maximum value* preclude separating this interference on an energy basis.

Although alpha and beta-gamma produced pulses are of different pulse-shape (or pulse-time-duration) in some liquid scintillator systems (13, 21), they cannot be separated on a pulse-shape basis using presently available commercial beta-liquid-scintillation equipment because the pulses are not processed in the optical or electronic system in such a way as to retain this information. Thus, although the total back-

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*In the case of beta radiation, because of the continuous beta energy distribution. In the case of gamma radiation, because of varying efficiency of gamma interaction with the scintillator.

ground is less with the extractive scintillator used in a commercial beta-liquid-scintillation counter (because ^{40}K and some other beta-gamma emitters are not extracted), the background from outside sources cannot be reduced much. Background counts of 15 to 20 counts/min are the usual lower limit.

Alpha spectroscopy with a high-resolution liquid scintillation detector

With appropriate detection and electronic equipment and a properly prepared sample, it is possible to obtain an alpha resolution of 200 to 300 keV FWHM. A background of about 1.0 count/min under an alpha peak can be obtained using a Pyrex tube to contain the counting sample; this can be reduced to 0.3 counts/min using a quartz tube. Backgrounds of 0.01 counts/min or lower are easily obtained when pulse-shape discrimination methods are used to eliminate beta-gamma pulses. Sample preparation methods for soil and water samples and the necessary equipment for high-resolution alpha spectrometry have been described earlier (16), as has pulse shape discrimination equipment for rejection of beta-gamma pulses (13).

Sample preparation for high-resolution liquid scintillation: Optimum pulse height, energy resolution, and pulse-shape discrimination require that the scintillator-sample contain a minimum of color quenchers or chemical quenchers. The extractant, the scintillator, and all other constituents of a high-resolution scintillator must be as pure as is practicable, the aqueous sample from which the activity is to be extracted must contain little or no nitrate or chloride (both chemical quenchers), and oxygen must be removed from the scintillator-sample. Methods for achieving such conditions are not as difficult as one might assume. In general, a single preliminary separation from gross impurities followed by extraction into the scintillator is sufficient for the purification. One such procedure using solvent extraction methods to isolate uranium and plutonium from soil and water samples has been described in a previous publication (16). Any of the precipitation, ion exchange, or solvent extraction methods that are used to separate alpha-emitting nuclides for analysis by surface barrier (or other) counting methods (22-24) can easily be adapted to isolation of the same nuclides for high-resolution liquid scintillation spectrometry.

An adaptation has been made in our laboratory of a chloride-system ion exchange procedure (24) for preliminary purification of some bone and tissue samples intended for plutonium analysis. The method consists of loading the sample (in a 10 M HCl, 0.01 M HNO_3 medium) onto a column of Dowex 2 x 8 that has prewashed with 10 M HCl, washing the

column with 10 M HCl and then stripping with 10 M HCl containing 1 ml of 45% HI per liter. Uranium is also retained by the column and may be removed after the plutonium by stripping with water. Of 11 spiked samples run by this method, the average recovery of plutonium was 87.5% with a standard deviation of 7.6%. Details of this method are being reported separately (25).

The procedure that we have found to be most convenient and most reliable for sample purification for plutonium analysis in bone and tissue samples is an adaptation of the solvent extraction method described earlier (16). After the sample has been placed in solution in nitric acid and the acidity adjusted to 1 to 2 M by evaporation and dilution, aluminum nitrate is added to saturation and the sample is extracted with an equal volume of 0.3 M high-molecular-weight tertiary amine (octyl, nonyl or decyl) nitrate in toluene.* Just prior to extraction, the plutonium valence is adjusted to (IV) by the addition of sodium nitrite. After extraction, the aqueous phase is discarded and the organic phase is washed once with 0.7 M nitric acid. The plutonium is then stripped from the organic phase with an equal volume of 0.3 M $HClO_4$ followed by two washes of 1/4 to 1/5 the organic-phase volume containing a total of 0.3 to 0.5 meq. of $HClO_4$ and about 30 meq. of $LiClO_4$. In the stripping step the 0.3 M amine nitrate is converted to amine perchlorate, releasing an equivalent amount of nitric acid to the aqueous phase; thus, the stripping solutions must contain sufficient perchloric acid for this exchange plus that intended to remain in the sample. The strip solution is then heated to a controlled temperature of 150 to 170°C in order to evaporate the nitric acid and reduce the volume while retaining perchloric acid (b.p. 200°C).** After the volume has been reduced to approximately 5 ml, the solution is diluted, while still warm, to 8 to 10 ml with a 2.5 w/v% solution of sodium peroxysulfate. This final solution should be approximately 3 M perchlorate and 0.05 to 0.1 M in perchloric acid with no other anions present. Plutonium recovery from solutions prepared as described has been 100% within counting statistics.

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*Amines containing more than 0.1 to 0.5% primary and secondary amines are not suitable (16).

**In the previous procedure (Ref. 16), fuming was continued until a lithium perchlorate fusion resulted. In some cases sufficient acid was removed to cause hydrolysis of the plutonium with attendant low recovery. The procedure described here is much superior.

For extraction into the scintillator, the sample is quantitatively transferred to a conical separatory funnel or centrifuge tube containing 1.2 to 1.5 ml of the high-resolution extractive scintillator composed of 4 g/liter of PBBO, 200 g/liter of naphthalene and 64 g/liter of HDEHP in toluene. After equilibration by manual shaking for 1 min and phase separation, a measured quantity of the scintillator is transferred to a 10 x 75 mm culture tube, placed in the high-resolution detector, and counted. The total plutonium count in the original sample is then calculated from the volume or weight ratio, scintillator taken/scintillator counted. Of 12 spiked samples reduced in volume, extracted, and counted in this way, the average plutonium recovery was 101% with a standard deviation of 2.4%.

The excellent plutonium recovery by these methods extends to low-level samples containing large amounts of unwanted ions. In six samples of approximately 130 g in size (~~1/2~~ of a rat), each containing a known 114 disintegration/min of plutonium, the average recovery was 112 counts/min or 98.4% with a standard deviation of 4.1%. No interference from naturally occurring radioactive elements was encountered. These samples were prepared by the acid dissolution, and furnace heating followed by amine solvent extraction procedure outlined for larger-sized samples under "sample preparation".

In all these procedures it is desirable that the volume of the aqueous phase in the final extraction be about 5 to 10 ml since the amount of plutonium extracted depends not only on the distribution coefficient of plutonium (concentration in organic phase/concentration in aqueous phase) but also on the volumes of each phase. It should also be emphasized that careful analytical technique with attention to quantitative transfers in all steps is required for good results.

When pulse-shape discrimination is to be used to reject the beta-gamma background, it is necessary to remove oxygen from the sample so that sufficient pulse-shape discrimination can be obtained. Previous publications have indicated that this would require freezing, evacuation, and helium or argon refilling of the sample tube (6, 16). Recent work has shown that excellent pulse-shape discrimination can be attained much more simply (17). Sparging with any of a number of inert gases, such as argon, carbon dioxide, or methane, for approximately 5 min produces a sample having excellent pulse-shape separation characteristics. Figure 4 shows beta-gamma and alpha pulse separation with such a sample and an illustration of the rejection of a beta-gamma background from an alpha spectrum. Samples prepared by this sparging method

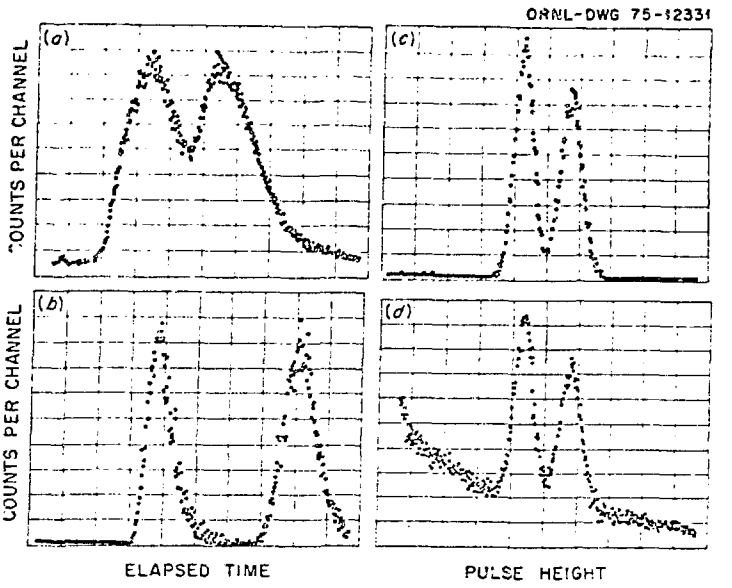

Fig. 4. Pulse-shape discrimination: Time spectra (a) before, (b) after argon bubbling; abscissa is 200 nsec full scale; β-γ pulses in left peak, α pulses right peak. Energy spectra (c) pulse-shape discriminator rejecting β-γ pulses, (d) without pulse-shape discriminator.

and closed with a cork covered with RTV silicon sealer usually remain stable and retain their good pulse-shape discrimination characteristics for more than 20 hr, some for many months.

Stripping from extractive scintillator for reanalysis in the high-resolution system: In some cases it has been found desirable to first analyze all samples by counting in the beta-liquid-scintillation counter using the extractive scintillator and then reanalyze those requiring better energy resolution or lower background with the high-resolution system. In order to do this, the plutonium must be stripped from the first extractive scintillator and reextracted into the high-resolution scintillator. The following procedure is used: The extracted sample is transferred from the 20-ml vial to a 60-ml separatory funnel using small volumes of toluene to wash. The aqueous phase is removed and discarded and the organic phase is washed twice with five-ml volumes of a solution that is 0.4 M in LiCl and 0.1 M in HCl (to remove nitrate from the organic phase). The washes are discarded. A 10-ml volume of 0.2 M solution of 2,5-di-tert-butylhydro-

quinone in 2-ethylhexanol is added to the separatory funnel [to reduce Pu(IV) and Pu(VI) to Pu(III)], and the plutonium is stripped from the organic phase with two 10 ml volumes of 6 M HCl. The combined HCl strip is then evaporated and prepared for extraction as described in the previous section. Eleven samples, each containing 1 gram of bone, treated in this way gave an average recovery of 97% of the plutonium added with a standard deviation of 1.7%.

CONCLUSIONS

Methods of alpha counting by liquid scintillation methods have been reviewed and compared and their successful application to counting plutonium in bone and tissue samples has been reported. A method of gross alpha counting using scintillators and equipment designed for beta scintillation counting in aqueous samples is shown to be useful where the alpha count rate is sufficiently high and where contribution to the background from beta or gamma emitters in the sample is sufficiently low. A second method in which beta-liquid-scintillation equipment is used but the plutonium is extracted into a water-immiscible scintillator was found to give more reproducible results and a lower background. For samples with low count rates or those requiring alpha energy resolution, an alpha-liquid-scintillation method is found to offer advantages of very low background (about 0.01 counts/min), the ability to selectively count alpha pulses while rejecting counts from beta or gamma emitters in the sample, and ~~a~~ energy resolution capability of 200 to 300 keV FWHM. Sample purification and separation methods based on solvent extraction (or on a combination of ion exchange and solvent extraction) for these alpha liquid scintillation methods are more simple and more rapid than those for most other alpha counting methods.

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