

**SOME APPLICATIONS OF
PHOTON/ELECTRON-REJECTING ALPHA LIQUID SCINTILLATION
(PERALS) SPECTROMETRY TO THE ASSAY OF ALPHA EMITTERS***

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ABSTRACT

The combination of certain solvent extraction separations and a special kind of liquid scintillation detector and electronics designed for alpha spectrometry allows some highly accurate, yet simple determinations of alpha-emitting nuclides. Counting efficiency is 99.68% with backgrounds of <0.02 cpm. Energy resolution and peak position are sufficient for the identification of many nuclides. Rejection of interference from β and γ radiation is >99.95%. The Photon/Electron Rejecting Alpha Liquid Scintillation (PERALS) equipment is described and procedures for the separation and determination of uranium, thorium, plutonium, polonium, radium, and trivalent actinides are outlined.

BACKGROUND

Many people have considered counting α -emitters by liquid scintillation methods(1-3). However, attractive as this idea is because of near 100% counting efficiency, no backscatter, no self-absorption, and simple methods of sample preparation, there are several non-trivial problems that often stand in the way of successful application of liquid scintillation to α -counting: (a) On a commercial β liquid scintillation (β -LS) spectrometer, the energy resolution is poor, usually ~1 MeV FWHM, and cannot be improved much. (b) The addition of a sample, particularly an aqueous sample, into a scintillator usually modifies the scintillator efficiency in such a way as to reduce the number of β - γ events detected and shift α spectra to lower positions on the energy-analog scale. This results in a non-reproducible energy scale and a nonpredictable background. In some cases the latter problem can be dealt with by integrating the α peak above the baseline defined by the β - γ background, but this

becomes more difficult as the relative α count becomes smaller and the α spectrum becomes more complex. (c) β and γ radiation within the sample or γ radiation from outside the sample produce light in the scintillator about 10 times more efficiently per KeV of deposited energy than do α particles. This results in a β - γ background under the α peak(s) that is high and not predictable and not easily overcome even by purification of the sample since many α -emitters have β and γ radiation either as part of their decay or of their daughters' decay.

About 20 years ago, the authors were faced with the problem of determining distribution coefficients between aqueous and organic phases for several trans-plutonium elements for the purpose of deriving the sulfate complex formation constants for these elements *via* computer fitting to a model. It was necessary that the numbers be accurate to $\pm 1.0\%$ or better for the computer program to derive meaningful formation constants. After determining that it was not possible to achieve this precision and accuracy by

plate-counting methods, we turned to liquid scintillation counting using a β -LS counter, based on a suggestion given to us by D.L. Horrocks, and found that under the right conditions it was possible to obtain the accuracy required. However, as we tried to expand this technique to other than single-nuclide, simple-matrix, laboratory-generated samples, the problems listed above appeared either singly or all together. Despite our conviction that liquid scintillation was a more accurate way to quantify α -emitters, it was not possible to do so with much reliability with many "Real World" samples.

At this juncture we again followed the work of Horrocks(4,5) and attempted to build a detector to give better resolution of α peaks in a liquid scintillation system. After trying many designs, we built a detector, the basic design of which is shown in Fig. 1, that had much improved energy resolution. This expanded our ability to separate α energies but not to deal with quenching or β - γ background.

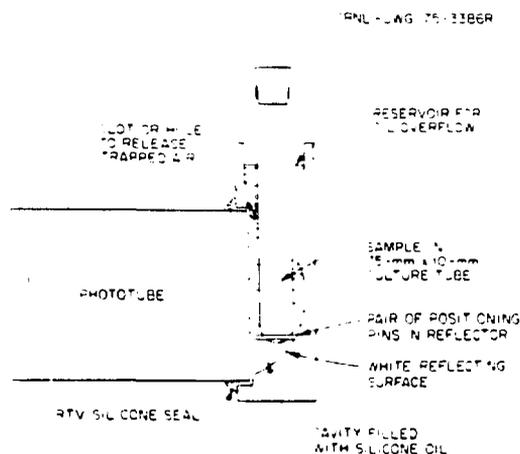


Fig 1. A Cross Section of the Detector/Reflector Assembly. The spherical cavity of the reflector is heavily coated with Eastman white reflectance coating. The cavity between the face of the phototube and the reflector is filled with silicone oil.

The research being done in our group was (and is) primarily concerned with metal ion separation using solvent extraction systems. Thus, the next step in improving α -liquid scintillation was natural. We developed scintillators that are also extractants, i.e., extractive scintillators. This allows a simple solvent extraction step to place the nuclide of interest in solution in the scintillator. This procedure also allows the scintillator to remain at reasonably constant composition even though the aqueous phase may vary somewhat in composition. Thus, the scintillator is of constant response to an α of a given energy, and the

α peak will appear at the same place on the energy scale for each sample. With this development we overcame problems a and b above. The composition of such scintillators is shown below.

PBBO, [2-(4'-biphenyl)-6-phenylbenzoxazole] - 4g
or α NPO, [2-(1-naphthyl-5-phenyloxazole)] -- 5g
Naphthalene, scintillation grade --- 160 to 200g
Extractant - 0.1 to 0.3 gram molecular weight
Toluene or *p*-xylene, distilled in glass, to make 1 liter.

Scintillator mixtures containing more than one fluor, e.g. a primary fluor and a wavelength shifter, do not perform as well in this system as those containing a single fluor(6).

Typical extractants that have proven useful are: cation exchangers such as:

bis(2-ethylhexyl)phosphoric acid (HDEHP),
high-molecular-weight (>250) neocarboxylic acids,
example, 2-heptyl-2-methylnonanoic acid (HMHN)

anion exchangers for example:

primary, secondary, and tertiary amines as sulfates
examples of which are:

1-nonyldecylamine sulfate (INDAS)
didecylamine sulfate (DDAS)
trioctylamine sulfate (TOAS)

and neutral coordinating extractants such as,
trioctylphosphine oxide (TOPO),
tributylphosphate (TBP),
various macrocyclic polyethers.

The "extractive scintillator" concept was an important step toward the practical use of liquid scintillation for α counting; however, the problem of a large and unpredictable β - γ background remained with us. The elimination of this problem is described below.

PULSE-SHAPE DISCRIMINATION

The use of pulse-shape discrimination (PSD) is well known(7,8) and is used in applications as divergent as neutron/gamma separation and "cleaning" recorded music of record surface noise. With the aid of John Thorngate, (9,10) we constructed a pulse-shape discrimination circuit that was able to sort the shorter β - and γ -produced pulses from the longer α -produced pulses and reject the β - γ pulses. No α pulses are lost. Figure 2 shows a block diagram of the set-up and Fig. 3 illustrates the steps involved in pulse-shape discrimination. The application of PSD to α -liquid-scintillation counting has allowed removal of the troublesome β - γ background in α spectra. Of the two circuit variations tried, timing of pulse rise time and timing of zero (or near zero) crossing on the descending portion of the pulse, it appears that the comparison of crossing of the falling integrated pulse gives the best results. There is more time

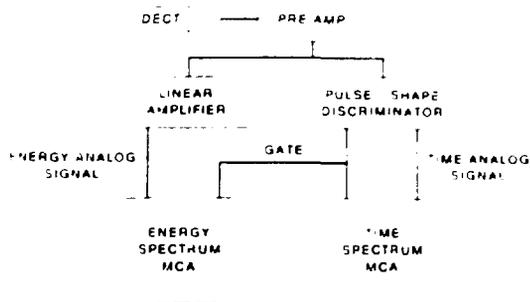


Fig. 2. A Block Diagram of the Pulse-Shape Discrimination Circuit. The ability to view the time spectrum allows one to be sure that all of the $\beta\text{-}\gamma$ pulses are rejected and all the α pulses retained; however, two MCA's are not required, an input-switching device is satisfactory. The use of two MCA's is simply a convenience.

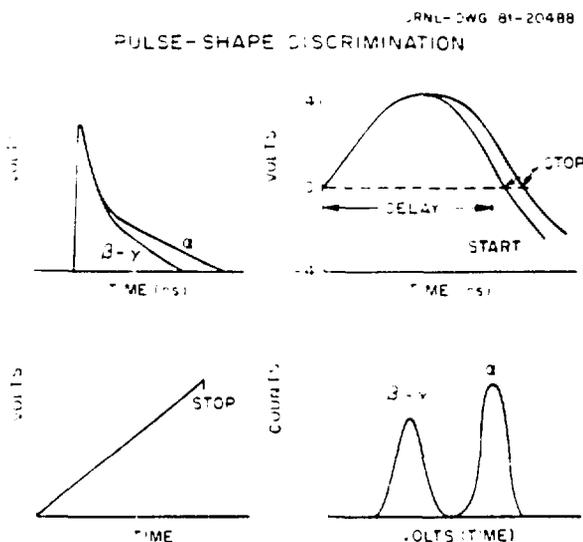


Fig. 3. The steps in Pulse-Shape Discrimination Shown Diagrammatically: top left, light pulse; top right, delay and placement of start and stop signals; bottom left, linear voltage ramp controlled by stop and start pulses; bottom right, typical time spectrum obtained. A logic pulse is sent to the MCA receiving the energy-analog pulses for every pulse occurring in the right peak. Thus, the MCA registers only α pulses.

information available at this point, 30 to 40 nanoseconds difference between α and $\beta\text{-}\gamma$ pulses, and the newer more stable delay circuits introduce little noise or inaccuracy in timing. Fig. 4 illustrates the effect on the spectra wrought by pulse-shape discrimination. It should be noted that PSD is possible only with a detector of the type shown in Fig. 1. Many attempts to employ PSD with other types of detectors, particularly with $\beta\text{-LS}$ counters, have failed, probably because of too many possible light paths from sample to photocathode.

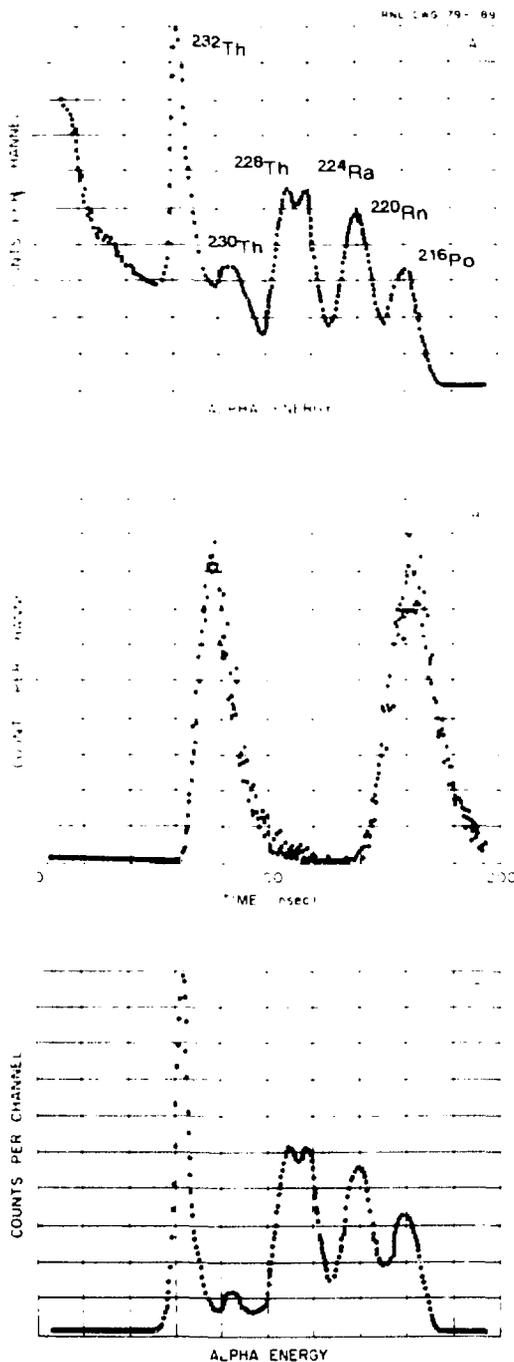


Fig. 4. Thorium-232 and Daughters; Top, with $\beta\text{-}\gamma$ Background; Middle, Time Spectrum; Bottom, With only α gated to the MCA.

At this point, a working system for obtaining useful α spectra from organic solutions in which the α -emitting nuclide is dissolved was at hand. Since the method rejected pulses produced by γ photons and β^+ or β^- particles, the complete system was named Photon/Electron-Rejecting Alpha Liquid Scintillation

(PERALS) spectrometry. Energy resolution was, and is, ~5% (200KeV at 4 MeV, 350 KeV at 7 MeV), rejection of β and γ pulses, 99.95%, background under a 5 MeV α peak, 0.02 cpm or less, α particle counting efficiency 99.68% and peak position is linear with α energy (Fig. 5) (11).

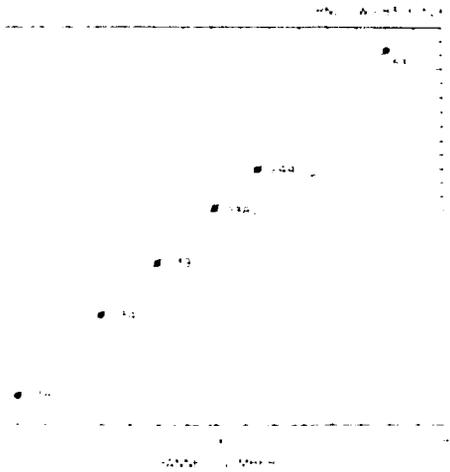


Fig. 5. Alpha Energy vs Peak Position. The same instrumentation settings and scintillator used in each determination.

REMAINING LIMITATIONS

From the above it might be possible for the reader to conclude that all problems were solved and PERALS was the perfect α assay method. This is nearly, but not quite, true. Certain limitations do apply, as they do in all analytical methods.

Quenchers

These can be colored materials that absorb light from the scintillation or absorb energy in a non-radiative manner. Both these processes move the α peaks to lower apparent energy but usually do not result in lost counts as in β -LS(12). However, there is usually a loss in both energy and pulse-shape resolution. Color quenchers can include colored organic compounds, humic acids, dyes, and colored metal ions. Because of the necessity of keeping color quenching to a minimum, all reagents and extractants must be as pure and color-free as possible. Energy or chemical quenchers are more insidious. Many colorless substances are severe quenchers. Organic and inorganic chlorides are serious quenchers as are alcohols and ketones. Nitrate and nitrite ions are both quenchers. Even dioxane, commonly used in aqueous-accepting scintillators is a quencher. Figure 6 shows the relative quenching of some extractants. Quenching is acceptable with TBP, TOPO, HDEHP, and the amine sulfates. Amine

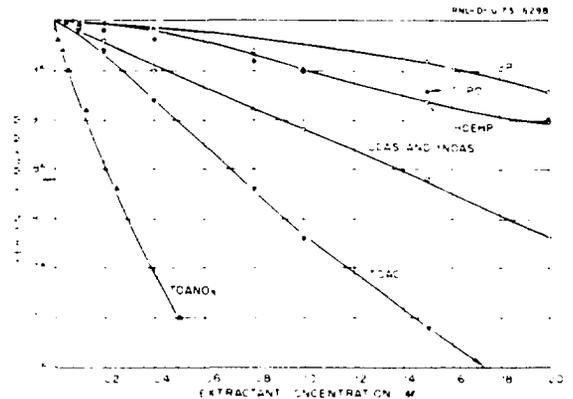


Fig. 6. The Quenching Effects of Several Commonly-Used Extractants. The abscissa is the percent of peak channel number in the presence of minimal extractant.

chlorides and nitrates, in general, quench too severely for practical use. However, The problem of quenching, in spite of the many things that quench, is not very limiting to the method. Enough excellent and selective extractants exist that do not seriously quench to solve most, if not all, problems. One last point before we leave quenching. Molecular oxygen is a quencher, and the best energy and pulse-shape resolution is obtained when it is sparged from the sample-containing scintillator. The method for doing this is quite simple. The 1.0-mL sample in the culture tube, ready for counting, is sparged for 1 to 2 min with an oxygen-free gas (such as argon, nitrogen, or methane) that has been saturated with toluene in a bubble tower. A 9-in Pasteur pipette is used as a sparging lance and discarded after use. The culture tube is stoppered tightly after sparging.

Resolution

Alpha energies that cannot be separated by 5% resolution present a problem. There are three approaches to this problem that have been successful. As was mentioned above, it is frequently possible to eliminate most interfering nuclides with the chemistry by which the sample is concentrated (preliminary separation) and introduced to the scintillator (selective extractive scintillator). Spectrum stripping programs of the type normally used for photometric spectra can be successfully applied to PERALS spectra because the curves are nearly Gaussian. There is some deviation from Gaussian due to the effect of conversion electrons (13), but this effect is not as large as theory would predict. The largest observed effect is <10% skew.

If in spite of the above, there may be peaks that overlap and are not identifiable. In this case, one may take a portion (or all) of the extractive scintillator, evaporate it on a plate, flame it, and

obtain a surface barrier spectrum. This spectrum will have superior energy resolution but inferior quantification of the total count. Thus, if one derives identity and ratio values from the surface-barrier spectrum and total count from PERALS, the resulting numbers for each nuclide should be more accurate than can be obtained from using either method alone under these circumstances.

Equipment Availability:

The PERALS detectors and PSD units have been individually custom built until very recently. Those used here were constructed in the Instruments and Controls Division of ORNL. Others were built at Brookhaven National Laboratory, Idaho National Engineering Laboratory, and in various universities. John McKlveen of Arizona State University and his students have contributed significantly to developing refinements for the construction and use of PERALS. The detector that evolved through all these efforts is shown in Fig. 7. At present, integrated, 3-nm wide units containing detector, PSD, and amplifier are being manufactured by a private electronics firm in Oak Ridge(14).

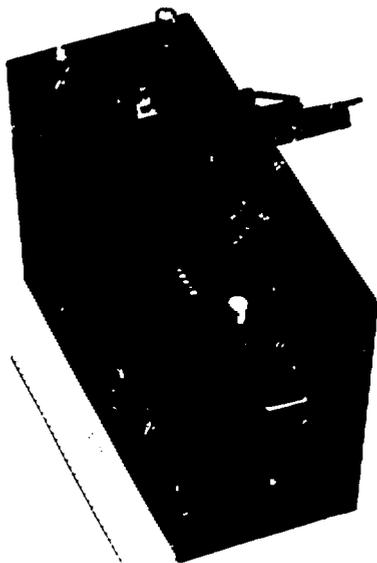


Fig. 7. Prototype PERALS Detector. The phototube and reflector are in a light-tight box with high voltage interlock switches. The connectors seen on top of the box are high voltage, interlock, and signal.

Improvements

Based on what we know at present, the critical items that allow improved energy and pulse-shape resolution include the phototube and reflector, the composition of the scintillator solution, and the matrix from which the sample is extracted. The phototube must have its maximum sensitivity in the region of light output by the fluors used. For PBBO this is 395 nanometers. The photocathode should be highly sensitive, i.e. release as many photoelectrons per α event as possible. At least one, and perhaps the largest, controlling factor in resolution is the number of electrons leaving the photocathode and arriving at the first dynode. Resolution is proportional to the square root of this number. The above being correct, it becomes important to collect and reflect as much light to the photocathode as possible. Many reflective materials were tested in the development of the PERALS detector, and a suspension of barium sulfate in alcohol-water with a small amount of polyester resin binder was found to be best among those tested(6,15). Specular reflectors were in general poor. The shape of the reflector and the sample size are both of considerable importance. The sensitivity of most photocathodes are quite nonuniform. Thus a highly reflective but diffusing spherical cavity reflector results in the best energy resolution because it distributes light uniformly over the photocathode with little dependence on the point of origin of the light(16). Hansche(16) demonstrated both mathematically and experimentally that this kind of reflector and a spherical cuvette of ~ 0.5 cm results in the best resolution. However, such a cuvette must be filled by vacuum distillation through a capillary and flame-sealed close to the sphere. Such a cuvette and method of filling is not amenable to practical use so a compromise of 1 ml of sample in a 10 X 75-mm culture tube was chosen. The loss of resolution with this arrangement, compared to the spherical cuvette, is small, $<1\%$.

The composition of the scintillator must be as non-quenching as possible and the aqueous phases with which the extractive scintillator is equilibrated must not transfer quenching materials to the organic phase. This means that substances such as organic-soluble dyes should not be in the aqueous phase and neither should it contain high concentrations of nitric or hydrochloric acids. Perchloric, phosphoric, and sulfuric acids and many salts do not extract and cause quenching in the organic phase.

At this point, it appears that the largest improvements in the performance of PERALS systems are likely to come from improvements in the phototube. A more efficient and a more uniform photocathode would very likely result in better energy resolution. A more

efficient fluor, one that produces more photons per alpha event, would also result in better energy resolution. We would welcome any suggestions on possible routes to such improvements.

PROCEDURES AND RESULTS

In general, the procedure for preparing samples for the PERALS spectrometry consists of: 1. Sample dissolution if needed. 2. Preliminary separation and/or concentration. 3. Transfer of the nuclide of interest to the scintillator by solvent extraction with the extractive scintillator. Both the preliminary separation and the extraction into the scintillator can serve as separation steps. These separations serve at least two purposes in addition to placing the nuclide of interest in the scintillator. 1. They remove the nuclide of interest from substances that interfere with the analysis, and 2. they increase the confidence that the α counts observed are from the desired element or group of elements.

Counting on Glass-Fiber Filters

Ordinary glass-fiber filters up to 2-inch diameter can be folded, inserted into a 10 X 75-mm culture tube, covered with HDEHP extractive scintillator, gently evacuated and released to argon two or three times to remove air trapped in the filter, and counted. This will work on relatively clean smear filters or air filters that are not too dirty. The spectrum obtained is not as good as that obtained from a nuclide in solution in an extractive scintillator. However, it is often good enough to allow identification of nuclides. If the filter or smear contains large amounts of dark-colored inert material this method cannot be used. In such cases the filter must be dissolved in an acid mixture containing hydrofluoric acid and extracted into a HDEHP-containing scintillator.

Uranium Activity on Cellulose Filters

Cellulose filters can be easily placed in solution and assayed as follows(17): The filter paper (up to 2-in diam.) is placed in a 2-dram, screw-cap borosilicate glass vial and heated, open, in an oven at 500°C for 2 hours. After removing and cooling the vial(s), 2 to 5 drops of concentrated nitric acid, 3 drops of 30% hydrogen peroxide, and 1 drop of a saturated aluminum sulfate solution are added to the vial. The vial is then heated to 200°C to remove the nitric acid. The solids remaining are redissolved in a solution 1.0 M in Na_2SO_4 and 0.01 M in H_2SO_4 . A measured quantity of an extractive scintillator containing a high-molecular-weight tertiary amine sulfate is then added to the vial and the two phases are equilibrated for 2 to 3 min. After the phases have separated, 1.0 mL of the organic phase is

counted on a PERALS spectrometer. By placing the vials in a rack or tray a number of samples can be processed at one time. Alpha spectra thus obtained usually allow identification of the isotopes of uranium present. Normal uranium in secular equilibrium will show the double peaks due to ^{238}U and ^{234}U . However, not all uranium now in the environment is normal uranium. If the uranium in the sample is highly enriched in ^{235}U , the spectrum will show primarily ^{234}U . Spectra of tailings from the enrichment processes will show a predominance of ^{238}U .

Gross Alpha in Environmental Materials

With the sample in solution in nitric or hydrochloric acid, 0.5 g of LiClO_4 and 1.0 to 1.5 mL of 0.1 M HClO_4 is added. The sample is then evaporated under heat lamps or in an aluminum block at 160°C until boiling of the sample ceases and the first perchloric acid fumes appear. Next, cool the beaker and add 5 to 7 mL of water to dissolve the viscous residue. Measure the pH of the solution; it must be between 2 and 3.5. Transfer the solution to a small equilibration vessel and add a measured quantity, 1.2 to 1.5 mL, of an extractive scintillator that is 0.2 M in HDEHP. Equilibration will transfer most of the α -emitting nuclides, with the exception of radium and radon, to the scintillator. Lanthanides are also extracted. Large amounts of these elements can cause incomplete extraction of actinides. Reequilibration of the aqueous from the first equilibration (after carefully removing all of the original organic phase) and counting this new organic will usually indicate if the first extraction was incomplete. Counting on a PERALS spectrometer will usually allow identification of the nuclides as well as an accurate quantification of them(18). If there are unresolved α peaks in the PERALS spectrum, evaporate a small portion of the extractive scintillator on a plate, flame it, and obtain a surface-barrier spectrum. Nuclide ratios obtained in this way can be applied to the total count on PERALS giving accurate total amounts of each nuclide(19).

Uranium and Thorium in Phosphates

These elements can be separated from a variety of phosphate-containing materials e.g., fertilizers, bones, teeth, animal tissues, and wastes(20,21). The sample is dissolved and placed in a nitrate or nitrate-perchlorate solution. To this solution, sufficient aluminum nitrate is added to complex the phosphate. This solution is then contacted with a toluene solution of trioctyl phosphine oxide (TOPO). Both uranium and thorium are transferred to the organic phase. The organic phase is then stripped with an equal volume of 0.5 M ammonium carbonate solution, the ammonium carbonate is evaporated, the sample is

converted to nitrate or nitrate/perchlorate, and any entrained organics are destroyed. The clear solution is then converted to a sulfate system by adding sulfuric acid and evaporating. The uranium is then extracted into a scintillator containing a tertiary amine sulfate such as trioctylamine sulfate, and the thorium is extracted separately into a scintillator containing a primary amine sulfate such as 1-nonyldecylamine sulfate. Each organic extractive scintillator solution is then sampled and counted on the PERALS spectrometer. *Note the statements on environmental uranium under "Uranium activity on Cellulose Filters".*

Polonium

The radioisotopes of polonium (usually ^{210}Po) have been difficult to analyze with accuracy. This procedure, however, is simple, rapid, and accurate(20). With the sample in solution in nitric acid or a mixture of nitric, hydrochloric, and hydrofluoric acids add ~5 mL of concentrated phosphoric acid and evaporate to remove other acids. If the original sample is in strong hydrochloric acid, nitric acid should be added and the sample heated before phosphoric acid is added. Otherwise, some loss of polonium by volatilization may occur. Transfer this phosphoric acid solution to a small equilibration vessel using about 5 mL of water. Add 1 mL of 0.1 M HCl. Add a known volume, 1.2 to 1.5 mL, of an extractive scintillator that is 0.1 M in TOPO. Equilibrate and count 1.0 mL on a PERALS spectrometer. Internal recovery tracers can be used. Because of the minimal chemical manipulations required, the accuracy of this determination is often better than $\pm 1\%$. Figure 8 illustrates spectra obtained in polonium analysis and the use of PSD to pull counts from a low-counting sample out of a high $\beta\text{-}\gamma$ background.

Plutonium

Plutonium can be chemically separated from all other elements except neptunium and counted quantitatively by this procedure and if ^{237}Np is present in amounts that interfere it can be separated from $^{239-240}\text{Pu}$ (21,22) by curve stripping. The initial extractant in this procedure is 0.3 M high-molecular-weight tertiary amine nitrate (TANO_3) in toluene. The sample should be in solution in 3 to 4 M $[\text{NO}_3^-]$ and 0.5 to 1.0 M HNO_3 . The plutonium is reduced with ferrous sulfate and reoxidized to Pu(IV) with sodium nitrite. This solution is equilibrated with not less than 1/4 its volume of TANO_3 solution. Separate the aqueous phase cleanly and wash the organic phase with two - 1/4-volume portions of 0.7 M HNO_3 . The aqueous from the first equilibration and the washes can be combined and analyzed for uranium, if desired. Plutonium can be

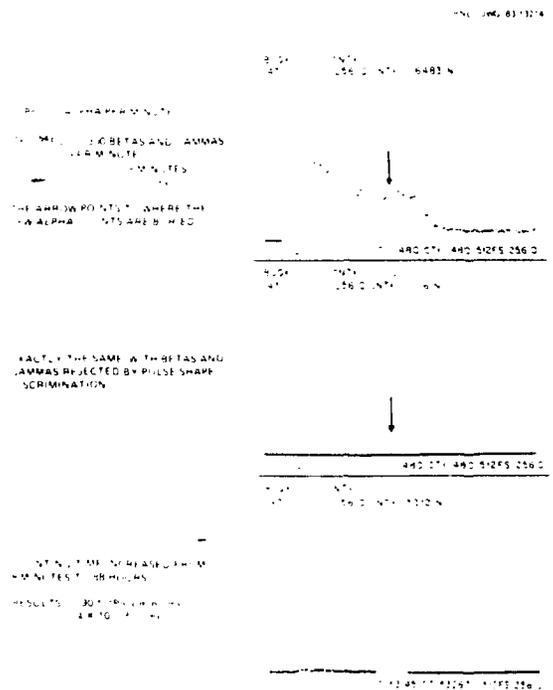


Fig. 8. Polonium counted; Top, in the Presence of a Large $^{152-154}\text{Eu}$ Background; Middle, Background Removed with PSD but Counting Time Not Increased; Bottom, PSD used and Counting Time Increased to 88 Hours.

stripped from the organic phase either with perchloric acid or, after diluting the organic phase with 2-ethylhexanol, with 1 N H_2SO_4 . The plutonium is extracted into a scintillator containing HDEHP in the first instance and into a scintillator containing 1-nonyldecylamine sulfate in the second instance.

Radium

A new extraction reagent allows radium to be separated from other alkaline earth elements, and the following procedure allows separation from many (if not all) other cations(23). The sample solution is first spiked with ^{133}Ba and 10 mg of barium carrier is added. Radium is then precipitated from the sample solution as barium/radium sulfate. The precipitate is now converted to the carbonate by heating with a saturated potassium carbonate solution, and the separated barium/radium carbonate is dissolved in dilute acid. The pH of this solution is then adjusted to between 9 and 10, and the radium is extracted selectively into an extractive scintillator containing a synergistic reagent mixture of a branched, high-molecular-weight carboxylic acid such as 2-methyl-2-heptylnonanoic acid (HMHN), 0.1 M, and dicyclohexano-21-crown-7 (DC21C7), 0.05 M. Table 1 shows separation factors obtained for several elements.

TABLE 1
RADIUM SEPARATION FACTORS

<u>RADIUM FROM</u>	<u>SEPARATION FACTOR</u>
Barium -----	9.3
Strontium -----	12.3
Calcium -----	>58
Magnesium -----	560 (Approx.)
Beryllium -----	2×10^6 (Approx.)
Zinc -----	7×10^6
Lithium -----	8×10^3 (Approx.)
Cesium -----	1.9×10^5
Thorium -----	3.7×10^6
Uranium -----	6.2×10^6

The extraction is quantitative, and any chemical losses in the coprecipitation and metathesis steps can be corrected by γ counting the ^{133}Ba . Radium is counted on a PERALS spectrometer, initially showing a single peak (Fig. 9). All α peaks from the radium daughters can be seen if one waits for their ingrowth (Fig. 10). The accuracy of this determination, with appropriate counting statistics, is usually better than $\pm 5\%$.

Strontium

Although this procedure does not use the PERALS electronics, it does use some of the technology, specifically the concept of the extractive scintillator.

The clean separation of strontium from environmental (and other) samples is almost as difficult as that of radium. We have developed a procedure for strontium that will separate it from many interfering elements; however, not all possible interferences have been examined. Beginning with the sample in 0.2 M HNO_3 many interferences are removed by extraction with 0.10 M HDEHP in toluene. The organic phase can be discarded or retained for the determination of U, Th, Np, Y, and several other elements. The aqueous phase is treated with a standard chromate precipitation(24). The liquid supernate is separated, made basic, and extracted with a scintillator that is 0.05 M in dicyclohexano-18-crown-6 (DC18C6) and 0.1 M in the sodium salt of either HDEHP or HMHN. The extract can be counted on a β -liquid-scintillation spectrometer. The difference in the β energy of the two strontium nuclides suggests that both ^{90}Sr and ^{89}Sr could be determined from a single count. Also, the

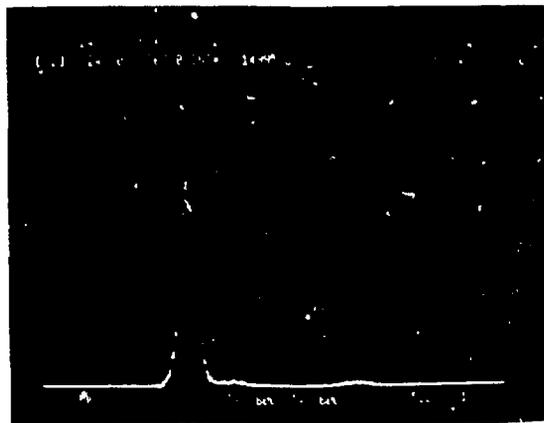


Fig. 9. Radium-226 Peak Counted 2-3 Minutes after extraction into the scintillator.

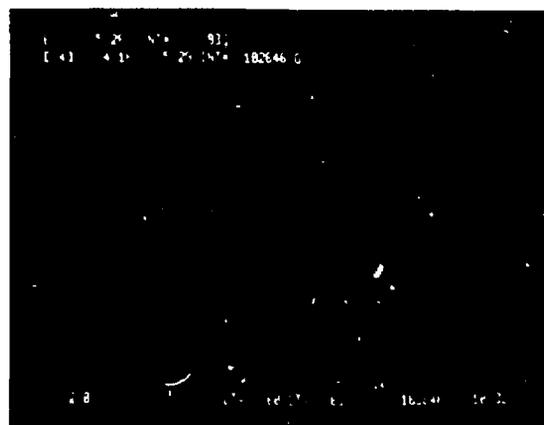


Fig. 10. Spectrum of Radium and Daughters 30 days after extraction and placing in a sealed counting tube.

ingrowth of ^{90}Y can also be followed as an additional check on the ^{90}Sr concentration, if desired(25). The use of the extractive scintillator in the last step eliminates most of the troublesome quenching problems associated with the addition of an aqueous solution to an aqueous-accepting scintillator and allows a more constant response to the β energies.

CONCLUSION

The success we have had with α analyses using the PERALS system over the last 20 years encourages us to propose that this method is a useful addition to the methods of α assay. In addition, the separation methods reported here may prove useful in other analytical or process chemistry applications. The PERALS system is complementary to the older methods, not a replacement. It has advantages in accurate quantification of α counts and ease of

sample preparation and disadvantages in energy resolution. The authors feel that with increasing use and familiarity with the method confidence in it will increase and its limitations will become better defined. We also feel certain that improvements in both equipment and supporting chemistry will expand the usefulness of PERALS in the future.

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