OPTIMIZATION AND CALIBRATION OF A LOW-BACKGROUND LIQUID SCINTILLATION COUNTER FOR THE SIMULTANEOUS DETERMINATION OF ALPHA AND BETA EMITTERS IN AQUEOUS SAMPLES

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ABSTRACT. The use of low-background liquid scintillation counting (LSC) techniques permits the simultaneous measurement of both alpha and beta emitters with efficiencies close to 100%. In this study, the counting parameters of a Wallac 1220™ Quantulus LS counter have been optimized for the best possible separation of the α and β spectra when measuring a sample containing 8 ml of water and 12 ml of Optiphase HiSafe™ 3 scintillation cocktail. For a normal quenching, minimum interference was observed when using an optimized α/β pulse-shape discrimination value, PSA = 117 (which ranges from 1 to 256). As the detection limits were increased when using a pulse-amplitude comparator to reduce the background, this circuit was not activated during the experiment. In these conditions, the total interference between the α and β spectrum was ~8%. This interference occurred only between the α window and the high-energy β window, and was little influenced by small variations of quenching. Better separation was achieved when the samples showed lower quenching. The figures of merit for the α and high-energy β windows were 144,000 and 1452 cpm^{-1}, respectively, corresponding to efficiencies of 95% and 77%. The minimum detectable activities for α and high-energy β emitters in water samples were ~0.15 Bq liter^{-1} and 1.36 Bq liter^{-1} for a total counting time of 360 min.

INTRODUCTION

Environmental studies, such as the determination of the quality of drinking water, often require measuring of mixtures of α and β emitters. The usual method for monitoring low-level α and β radioactivity is to use gas-proportional detectors where the ionization and proportional regions are used for α and β counting, respectively. In practice, the usual efficiencies for such a system are about 15% for α and 30% for β emitters, due to geometry limitations and absorption in the detector windows. For continuous-flow proportional detectors, solid scintillation (ZnS screens or plastic scintillators) or semiconductor detectors (surface-barrier or ion-implanted detectors), the maximum efficiency is <50%, due to geometry limitations. There are two problems with these techniques: 1) the determination of both α and β emitters requires two independent analyses; 2) given that the efficiencies are relatively low, counting times must be extended to obtain lower detection limits and/or better statistics.

With liquid scintillation counting (LSC), counting times are reduced because efficiencies are closer to 100%. Further, it is possible to measure simultaneously both α- and β-emitting radionuclides because their pulses can be identified using pulse-shape analysis (PSA). We have studied the simultaneous determination of α and β emitters in aqueous samples using a Wallac 1220™ Quantulus low-background LS counter.

METHODS

In a Quantulus 1220™, background is reduced by using an asymmetric passive (lead, cadmium and copper) shield, an asymmetric active (LS) shield and low-activity materials. The PSA and pulse-amplitude comparator (PAC) circuits are also accesssible to the user. We have optimized the pulse-discrimination capabilities of the detector to achieve the best α/β separation and the lowest detection limits possible. For all measurements, we used polyethylene vials (Wallac) and Optiphase

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HiSafe™ 3 (Wallac) scintillation cocktail. Although low-background Teflon-copper vials may reduce considerably the detection limits, we considered them too expensive for routine measurements.

The optimization and calibration of the Quantulus 1220™ included the following:

1. Optimization of the discrimination of α and β pulses using the PSA circuit
2. Optimization of the rejection of background signals based on the comparison of their amplitudes using the PAC circuit
3. Optimization of the sample/scintillant ratio
4. Efficiency calibration of the detector
5. Energy calibration of the detector
6. Determination of the instrumental background and detection limits.

RESULTS

PSA Optimization

The discrimination of the α and β pulses is based on the well-known difference between the delayed component of their fluorescence decay (Bollinger 1961; Knoll 1989). The Wallac PSA compares the area of the delayed component of the pulse with its total area and discriminates the pulses using a software-controlled parameter that varies from 1 to 256 (Kaihola 1990b). The pulses identified as α events are analyzed and routed to a multichannel analyzer (MCA), called α spectrum. The rest are stored in a different MCA, called β spectrum, which was divided into three regions of interest:

1. The low-energy (or ^{3}H) window: channels 1–200
2. The medium-energy (or ^{14}C) window: channels 200–500
3. The high-energy (or ^{40}K) window: channels 500–1000.

We observed little interference (<2%) between the α and the low- and medium-energy β windows, although the high-energy window was affected by α pulses. Let α be the count rate in the α window and β the count rate in the high-energy β window. We defined α interference (τ_α) as the fraction of α particles observed in the high-energy β window when measuring an α emitter

$$\tau_\alpha = \frac{\beta}{\alpha+\beta} \quad . \tag{1}$$

We defined β interference (τ_β) as the fraction of high-energy β pulses observed in the α spectrum when measuring a β emitter

$$\tau_\beta = \frac{\alpha}{\alpha+\beta} \quad . \tag{2}$$

Finally, we defined the total interference as the sum of both

$$\tau = \tau_\alpha + \tau_\beta \quad . \tag{3}$$

The optimization of the PSA parameter was made by minimizing the total interference. We measured separately an α emitter (^{241}Am) and a high-energy β emitter (^{32}P) using 8 ml of water and 12 ml of the scintillation cocktail. We also studied the effect of quenching on the pulse-shape discrimination by adding small quantities of CCl_4 to the scintillation cocktail. The quenching was monitored by the sample quenching parameter (SQP(E)); this corresponds to the channel below which lies 99% of a ^{226}Ra Compton spectrum (Rundt 1989).

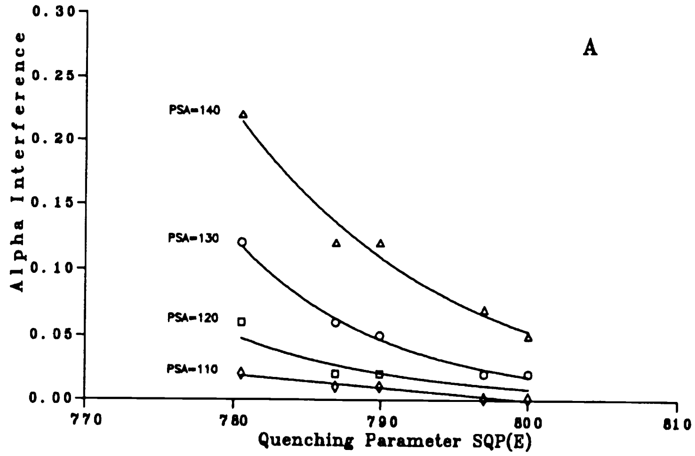

Fig. 1A. α interference as a function of quenching for different pulse-shape discrimination values

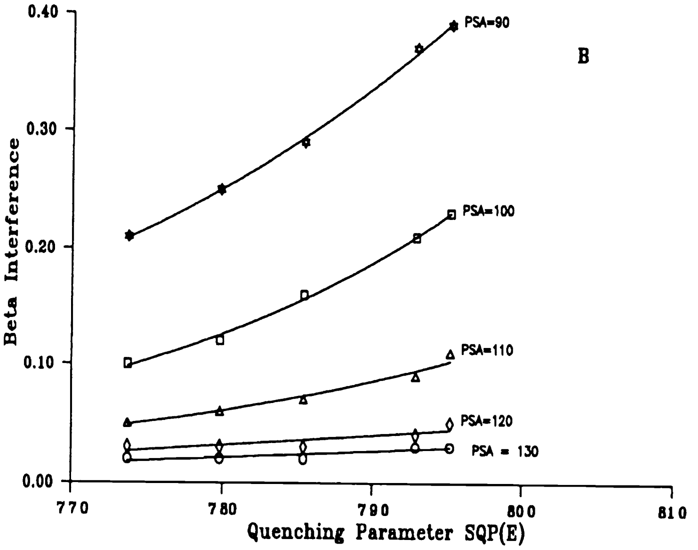

Fig. 1B. β interference as a function of quenching for different pulse-shape discrimination values

We observed the zone of minimum total interference for pulse-shape discrimination values, ranging from 90 to 140. For fixed quenching, we observed that an increase in the pulse-shape discrimination value caused the α interference to increase and the β interference to decrease, as expected (Fig. 1). The total interference-PSA distributions were well fitted with exponential curves. Fig. 2 shows that the distribution of the total interference as a function of quenching was well described by quadratic curves, thus, finally obtaining a complete description of the function, τ = f (PSA, SQP(E)), as Figure 3 illustrates. We observed:

1. The minimum total interference for the experimental conditions described was *ca.* 8%. This amount must be taken into account in the analyses.
2. For a quenching parameter, SQP(E) = 782 (average value observed in water samples), the minimum interference occurred for a pulse-shape discrimination parameter, PSA = 117. However, this value is NOT critical, as the τ function is almost independent of quenching and pulse-shape discrimination in that area.
3. It is almost impossible to obtain exact uniformity of quenching values in environmental samples. However, if the pulse-shape discrimination value was correctly chosen, variations of ten quenching parameter units produced a change in the total interference of only 1%.

4. We noted better separation of α and β pulses for lower quenching and, therefore, a higher SQP(E) parameter.

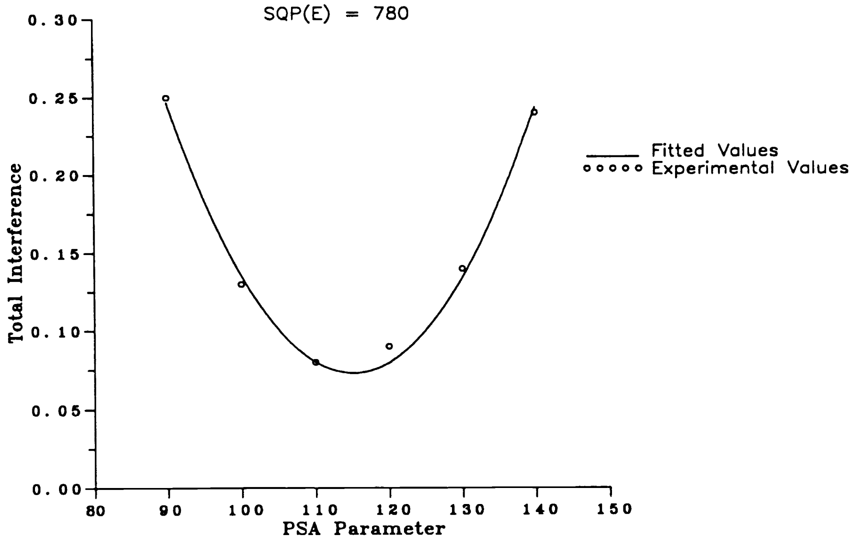

Fig. 2. Total interference as a function of pulse-shape discrimination for a given quenching

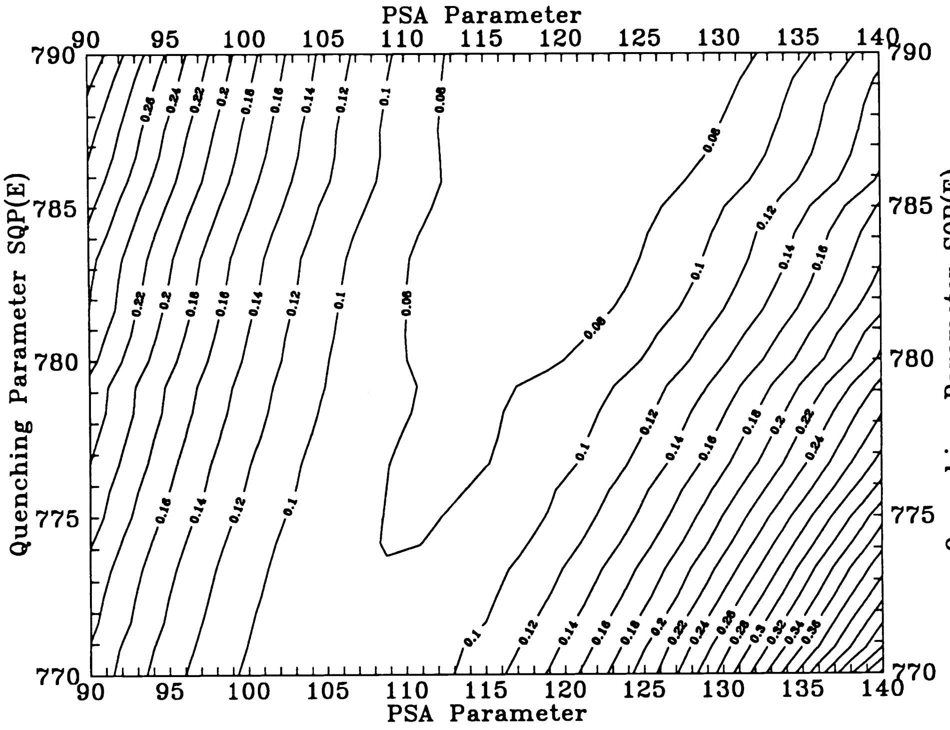

Fig. 3. Total interference as a function of quenching and pulse-shape discrimination

PAC Optimization

External radiation can cause Cerenkov or scintillation phenomena in one of the photomultiplier tubes (PMT). Thus, the optical crosstalk between the PMTs can increase the detector background. However, these coincident pulses show a smaller/greater amplitude ratio close to zero and can be rejected by the PAC circuit by choosing an adequate threshold. This circuit is controlled by a parameter that ranges from 0 to 0.8 smaller/greater ratio (1–256 in the instrument software).

We minimized the figure of merit (FM, cpm^{-1}) as a function of the pulse-amplitude ratio by measuring the activity of different β emitters (^{3}H, ^{14}C and ^{40}K) and the background with increasing quenching. The observed values (Fig. 4) showed that the PAC technique did not affect the FM in the low-energy window, but the efficiency was slightly reduced, thus reducing throughput, and that the FM was reduced in the medium- and high-energy windows as the PAC parameter increased. Thus, we decided not to use the PAC technique to reduce background. Other researchers observed a similar behavior when using polyethylene vials (Oikari 1987; Kaihola 1990a).

Optimization of the Sample/Scintillant Ratio

To place the maximum amount of sample in the vial, we determined the optimum sample/scintillant ratio using the total available volume of 20 ml. For this, we calculated the minimum detectable activity (MDA) (Currie 1968) for different proportions of tritiated water and the scintillation cocktail, assuming that a similar result should be obtained when using other β or α emitters.

As shown in Figure 5, the optimum sample volume was *ca.* 9 ml, which is about the highest holding capacitiy of the Optiphase HiSafe™ 3 scintillation cocktail for distilled water (Kaihola 1990b). However, as environmental water samples may induce phase separation, we decided to use the ratio, 8/12.

Efficiency Calibration

We determined the detector efficiency in the conditions used here (PSA = 117, SQP(E) ≈ 782, sample volume 8 ml, scintillation cocktail (Optiphase HiSafe™ 3) 12 ml) using ^{209}Po, ^{3}H, ^{14}C and ^{40}K standards. To study the variation of efficiency with quenching, we added aliquots of CCl_4 to the β standards. The observed β efficiencies (Table 1) increased with energy and decreased with quenching. For high-energy β emitters, the efficiency at quenching SQP(E) = 782 was 77.3 ± 1.5%. For α emitters, the efficiency was 95.1 ± 1.8 %.

Energy Calibration

We observed good logarithmic behavior of the energy-channel calibration for both α and β emitters (Pujol 1992). As the Quantulus 1220™ MCA channel scale is logarithmic, the overall dependence of the pulse intensity with energy was log–log. The β maximum energy-channel calibration was

$$\ln E_m\,(\mathrm{keV}) = (1.07 \pm 0.19) + (7.72 \pm 0.31) \times 10^{-3} \times \mathrm{channel}$$

and the α energy-channel calibration was

$$\ln E\,(\mathrm{MeV}) = (-0.62 \pm 0.13) + (3.68 \pm 0.20) \times 10^{-3} \times \mathrm{channel}\,.$$

Both calibrations showed regression coefficients, $r > 0.99$, and overall statistical significances, $F > 99.9\%$.

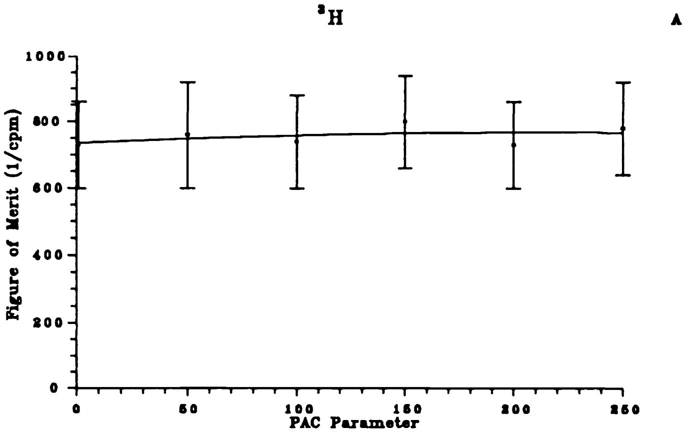

Fig. 4A. Figure of merit as a function of pulse-amplitude comparison discrimination when measuring ^{3}H

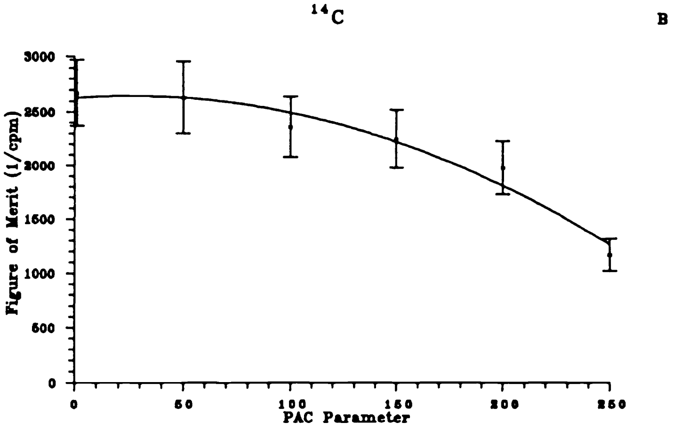

Fig. 4B. Figure of merit as a function of pulse-amplitude comparison discrimination when measuring ^{14}C

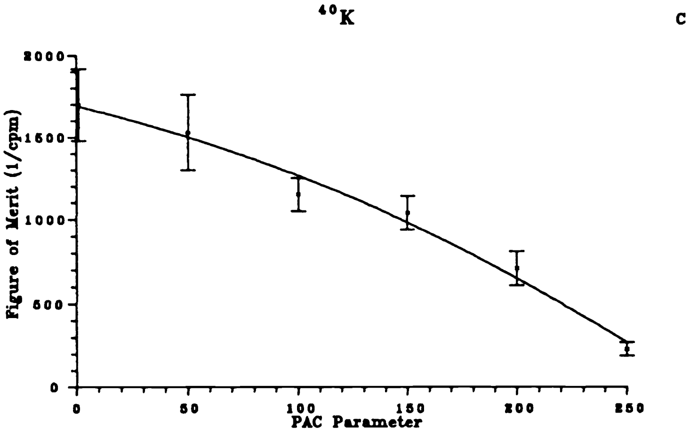

Fig. 4C. Figure of merit as a function of pulse-amplitude comparison discrimination when measuring ^{40}K

Background and Detection Limits

The number of significant sources of background in the detector system is large (scintillation cocktail, vial, PMTs, surrounding materials, cosmic rays, *etc.*). To determine a realistic background for low-level counting, we used two criteria: 1) low-energy window (1–200): we used very old groundwater, with no ^{3}H present, as the best available sample background; 2) medium- (200–500)

TABLE 1. Counting Characteristics of a Quantulus 1220™ Liquid Scintillation Detector System*

Window	Background (cpm)	Efficiency (%)	FM (cpm^{-1})	MDA** (Bq $liter^{-1}$)
1–200 (^3H)	1.466 ± 0.067	26.6 ± 1.2	483 ± 23	2.38
200–500 (^{14}C)	2.518 ± 0.066	60.0 ± 1.5	1430 ± 81	1.36
500–1000 (^{40}K)	4.115 ± 0.088	77.3 ± 1.5	1452 ± 64	1.36
400–800 (α)	0.063 ± 0.013	95.1 ± 1.8	(144 ± 30) × 10^{-3}	0.15

*Reported uncertainties = ± 1 σ

**Minimum detectable activity; counting time = 360 min

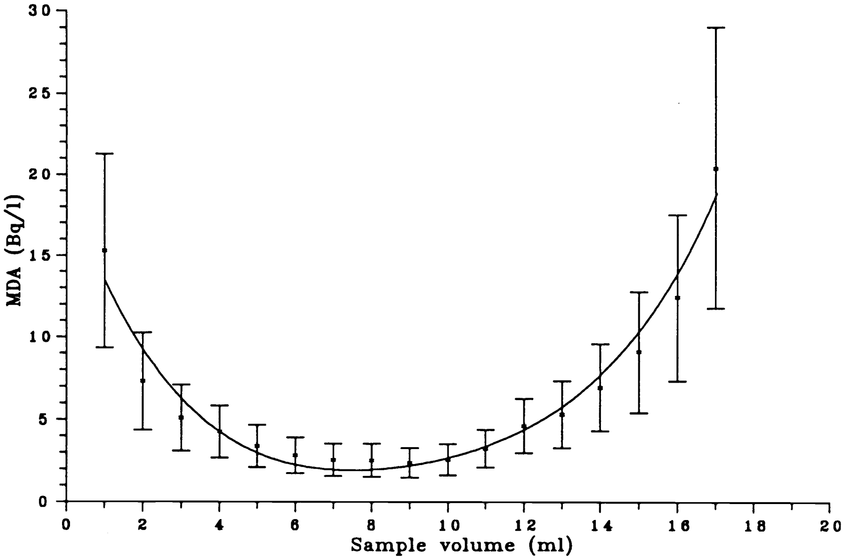

Fig. 5. MDA as a function of sample volume (total volume is kept at 20 ml)

and high-energy (500–1000) windows: as no such "old water" exists, background was considered to be proportional to the volume of scintillant used. We measured the background count rate per unit volume of scintillant using 20 ml of Optiphase HiSafe™ 3 to reproduce the counting geometry. In all cases, we quenched three background samples by adding different amounts of CCl_4 and obtained the background at any quenching by linear interpolation.

Table 1 shows the figures of merit for the defined windows. The observed values were high, as the windows were NOT optimized for each radionuclide, and normal polyethylene vials were used. When measuring water samples for the determination of total-α and total-β activities, we observed that the MDAs were 0.15 Bq $liter^{-1}$ and 1.36 Bq $liter^{-1}$, respectively, for a counting time of 360 min. Further, the World Health Organization (WHO) "action levels" for total-α and total-β activities in drinking water are 0.1 Bq $liter^{-1}$ and 0.8 Bq $liter^{-1}$, respectively (WHO 1979). To achieve an MDA equal to the WHO action levels, counting times of *ca.* 1000 min are required.

CONCLUSIONS

The usual method for monitoring low-level α and β radioactivity is to use gas-proportional counting, solid scintillation or semiconductor detectors. The use of LS techniques are a good alternative, as the determination can be done simultaneously by using pulse-shape discrimination techniques, and the efficiencies are closer to 100%, thus reducing counting times.

In this study, we optimized a low-background Quantulus 1220™ detector system for the simultaneous determination of α and β emitters in aqueous samples, and conclude that:

1. The minimum total interference between the α and high-energy β windows was about 8%. This value must be taken into account in the analyses.
2. The interference between the α window and the low- and medium-energy windows was negligible.
3. The minimum total interference was observed for a pulse-shape discrimination parameter of 117. This value is NOT critical, as the interference was almost independent of the parameter in the range of quenching studied.
4. If the PSA parameter was correctly chosen, small variations of quenching produced little change in the total interference.
5. We observed better α- and β-pulse separation for lower quenching.
6. The use of PAC for the rejection of background pulses did not improve the detection characteristics of the system when using polyethylene vials.
7. The optimum sample/scintillant ratio was about 9/11 ml. Taking into account the holding capacity of the scintillation cocktail, we chose a ratio of 8/12 for environmental samples.
8. The energy calibration of the detector showed a log–log overall behavior.
9. The efficiency of the detector was 95% for α emitters and 77% for high-energy β emitters. As the background of the detector was very low, the MDAs for a counting time of 360 min were only 0.15 Bq liter^{-1} and 1.36 Bq liter^{-1} for α and β emitters, respectively.

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