Mixed waste reduction in radioactivity determination by using plastic scintillators

A. Tarancon, J.F. García*, G. Rauret

Departament de Química Analítica, Universitat de Barcelona, Diagonal 647, E-08028 Barcelona, Spain

Received 4 January 2002; accepted 23 April 2002

Abstract

In this work, we tested whether plastic scintillation (PS) is a suitable alternative to liquid scintillation (LS) and Cerenkov techniques for beta emitter detection. The main advantage of this alternative is the reduction of mixed waste produced as consequence of the measurement process. In addition, the quality parameters obtained with PS are as reliable as those obtained with LS and allow determination, with a relative error <10%, for 90 Sr/90 Y in low level activity aqueous samples. Gamma and alpha emitters were also measured with plastic scintillators using linear and logarithmic amplified scintillation detectors.

Keywords: Radioactivity; Mixed waste; Plastic scintillators; Liquid scintillator; Cerenkov

1. Introduction

The wastes produced by nuclear activities are classified according to their chemical and radionuclide composition. Among them, those containing radioactive and organic compounds (mixed wastes) are difficult to dispose of because of the regulations established for nuclear and hazardous wastes [1] (in the US, these regulations depend on the Nuclear Regulatory Commission (NRC) and the Environmental Protection Agency (EPA)).

Although their activity can be low, the large amount of mixed wastes [2] makes them difficult to manage. Mixed wastes originate in the manufacture of labelled chemicals [3] and, mainly, in the emulsions generated in beta emitter determinations by liquid scintillation (LS) techniques in biomedical [4] and environmental studies [5]. In these emulsions, the organic waste results from mixing LS cocktails and aqueous radioactive samples. Therefore, the use of plastic scintillators instead of liquid cocktails may facilitate the segregation, after the measurement, of sample and scintillator without introducing additional waste in the measurement step.

The use of plastic scintillators is currently restricted to a few commercial products like filters in biochemical applications [6,7], and packing [8] in LS detectors coupled to liquid chromatography [9,10]. Comparing LS versus plastic scintillation (PS) characteristics for radionuclide determination [11,12], the former has slightly higher detection efficiencies, whereas the latter does not produce mixed wastes and it has the potential advantage to be reused after cleaning [13,14].

We aimed to determine whether plastic scintillators can be used as an alternative to LS cocktails for beta emitter determination. The performances of both were compared for the determination of a beta 90 Sr/90 Y in secular equilibrium, a low-energy beta emitter, 3H, 0003-2670/02/$ – see front matter © 2002 Elsevier Science B.V. All rights reserved.

PII: S0003-2670(02)00352-5
an alpha emitter, $^{238}$Pu, and a beta + gamma emitter, $^{134}$Cs.

2. Experimental

2.1. Reagents and solutions

All reagents used were of reagent or analytical grade. The active stock solutions used for the preparation of calibration and test solutions were as follows.

(a) Calibrant of 31.15 ± 0.46 M disintegration per minute per gram (Mdpn/mg) in 0.1 M HCl from Amersham International.

(b) S1: $^{90}$Sr/$^{90}$Y: 511.99 ± 7.67 dpm/g prepared from solution (a).

(c) S2: $^{90}$Sr/$^{90}$Y: 52.84 ± 0.79 dpm/g prepared from solution (a).

(d) S3: $^{90}$Sr/$^{90}$Y: 5.31 ± 0.08 dpm/g prepared from solution (a). In all these calibrant solutions, $^{90}$Sr/$^{90}$Y are in secular equilibrium, although nominal activity was related to the $^{90}$Sr activity.

(e) $^3$H: 5424.7 ± 108.49 dpm/g (prepared from a calibrant of 4.69 ± 0.09 Mdpn/mg in water from Amersham International).

(f) $^{134}$Cs: 1987.81 ± 9.94 dpm/g (prepared from a calibrant of 226.80 ± 1.34 dpm/g in water from Amersham International).

(g) $^{238}$Pu: 166.93 ± 0.83 dpm/g (prepared from a calibrant of 5005.81 ± 23.26 dpm/g in 2M nitric acid from Centro de Investigaciones Energéticas Medioambientales y Tecnológicas (CIEMAT)).

All the active solutions were prepared by diluting a weighed amount of standard solution in a weighed amount of a specific medium. Distilled water was used for the preparation of the calibration solutions.

Five water samples were used for the preparation of the test solutions. These samples were from the Rubí river (M3), the Martorell purifying plant (M4), the Mediterranean sea in front of the Vandelles I nuclear power plant (AM), underground water from the neighbourhood of the Ascó nuclear plant (Spain) (AS) and commercial bottled drinking water Fontvella® (EB). All the sampling points are located in Catalonia, NE Spain.

Blanks were prepared with inactive water. The quenching agent was—Bromocresol green (0.8, 0.1, 0.05 g/l). For LS measurements, the scintillation cocktail used was Optiphase-Hisafe2® (from EGG–Wallac) and the vials used were 7 ml polyethylene from Packard Instruments Co. For PS measurements, solid plastic scintillator beads of BC-400 in polyvinyl toluene were used instead of the Scintillation cocktail mentioned above. The diameter of the beads was between 250 and 500 µm. Vials used were made of plastic scintillator BC-408 in polyvinyl toluene. The shape of these vials was equivalent to that of the 7 ml polyethylene vial. Both the vials and the beads were supplied by Bicron.

2.2. Apparatus

A Tri-Carb 2000 CA/LL LS detector (Packard) with linear amplification, a multichannel analyser of 4096 channels (0–2000 keV) and background reduction based on pulse-shape discrimination were used. A Quantulus LS detector (EGG–Wallac) with logarithmic amplification, a multichannel analyser of 4096 channels distributed in four segments of 1024 channels, alpha/beta discrimination and background reduction by active guard were used.

2.3. Procedure

2.3.1. Measurements and solution preparation

For Cerenkov measurements, five calibrant solutions were prepared by adding 0.3 ml of the $^{90}$Sr/$^{90}$Y active solution (S1), 0, 1, 2, 3 or 10 drops of 0.1M bromocresol green and distilled water up to 3 ml in a 7 ml polyethylene vial. The activity in the vials was between 160 and 165 dpm.

For LS, six calibrant solutions were prepared by adding 0.3 ml of the $^{90}$Sr/$^{90}$Y active solution (S1), 0, 1, 2, 3 or 10 drops of 0.1M bromocresol green and distilled water up to 3 ml in a 7 ml polyethylene vial. The total activity in the vial was between 159 and 164 dpm. The amount of each fraction was determined by weighting.

For PS measurements, six calibrant solutions were prepared for $^{90}$Sr/$^{90}$Y determination by adding to the plastic scintillator vials 0.425 g of plastic scintillator beads, bromocresol green (four vials with 0–3 drops of 0.05 g/l and two vials with 3 drops of 0.1 g/l),
0.224 ml of the S1 active stock solution and distilled water to 0.5 ml. Single calibrant solutions were prepared for $^3$H, $^{134}$Cs and $^{238}$Pu following the same procedure without quenching agent addition. The activity in the vials was between 40 and 60 dpm for $^{90}$Sr/$^{90}$Y measurements, 500 dpm for $^3$H, 20 dpm for $^{238}$Pu and 215 dpm for $^{134}$Cs. For $^{90}$Sr/$^{90}$Y, two sets of three solutions each were prepared for limit detection evaluation following the previous procedure. The matrix of these samples was underground water and the total activity was 23.1 and 2.4 dpm/g for each set. S2 and S3 active stock solutions were used, respectively.

For $^{90}$Sr/$^{90}$Y determinations, one blank was prepared for each active calibration or test solution except for Cerenkov measurements, in which two solutions of extreme quenching were prepared. In all cases, active fractions were replaced by 0.1 M HCl when $^{90}$Sr/$^{90}$Y was determined, by 2 M HNO$_3$ for $^{238}$Pu and inactive distilled water for $^3$H and $^{134}$Cs. In test solutions, distilled water fractions were replaced by the water samples studied (M3, M4, AM, AS and EB) and no quenching agent was added. Once prepared, the vials were shaken and left to stand in the dark for 2 h before measurement.

2.3.2. Cleaning procedure
Vials and beads of plastic scintillator were reused. To remove the remaining activity, a specific cleaning procedure was applied for each isotope.

For $^{134}$Cs and $^{238}$Pu measurements, the protocol followed was:
1. rinse the vials with 0.1 M HCl;
2. shake the vials for 15 min in a 250 ml polyethylene bottle filled with 0.1 M HCl;
3. rinse the vial with distilled water;
4. apply an ultrasonic bath for 15 min to the vials in a 250 ml polyethylene bottle filled with distilled water;
5. as step 2, but using non-ionic soap solution (Count-Off®) for 12 h;
6. as step 5, but using distilled water; and
7. dry the vials using a paper filter.

The protocol for the beads was the same, but an additional filtration step with a Buchner and a Kitasato was included whenever the cleaning solution was changed. In step 7, the beads were dried in a heater at 40 °C.

For $^{90}$Sr/$^{90}$Y and $^3$H measurements, the protocol for vials and beads used was the same as described above but without step 5, related to the non-ionic soap solution bath. After each cleaning procedure, a blank was prepared using cleaned vials and beads to check that the activity was removed and both beads and vials could be reused.

2.3.3. Measurement step
Measurement was performed under the optimum conditions for each radionuclide. In the Packard Tri-Carb 2000 detector, all measurements, except the $^{90}$Sr/$^{90}$Y Cerenkov ones, were made with the low level (LL) option off. In the Quantulus detector, for $^{90}$Sr/$^{90}$Y and $^3$H measurements, the bias (CB) was low and the beta energy (BE) was high, whereas for $^{238}$Pu and $^{134}$Cs, CB was low and BE was high. In all cases, measurement time consisted of five successive periods of 1 h.

2.3.4. Quenching correction and data treatment
The five spectra collected for each solution were smoothed using a Savitzky–Golay algorithm and averaged. Activity was calculated by using the optimum window (OW) for each isotope and counting mode. The OW was established by using the unquenched calibration solutions; it corresponds to the range of spectra with the best figure of merit (FM = $E^2/B$, where $E$ is the efficiency and $B$ the background). For single measurements, theoretical uncertainty was calculated, whereas experimental standard deviation was used for replicate samples.

For each solution, the sample channel ratio (SCR) parameter was calculated as the ratio of the counting rates in two windows of the averaged spectra. These two windows were established, for each isotope and counting mode, as those in which the difference between the SCR of the extreme quenched solutions was higher.

Transformed spectral index of the external standard (T-Se) parameter is defined by Packard and is related to the position of the spectra obtained when the measurement solution is irradiated with an external gamma source.

Quenching correction curves were obtained by fitting OW detection efficiencies of the calibration
solutions versus SCR and T-SiE. For test solutions, the activity was calculated as the ratio of the integrated signal of the OW and the detection efficiency, which was obtained by interpolation of the quenching parameter value on the corresponding quenching correction curve.

The theoretical detection limit was calculated according to the HALS criterion [15]:

$$DL = 4.66 \times s(B)/(W \times E),$$

where $s(B)$ is the standard deviation of a blank with a quenching parameter value similar to that of the test solution value, $W$ the weight of the sample, and $E$ is the efficiency of an active calibration solution with a quenching parameter value similar to the test solution value.

2.4. Safety considerations

All the experimental procedures were performed in accordance with the regulations of the Spanish Nuclear Authorities (Consejo Seguridad Nacional) and the University of Barcelona.

3. Results and discussion

The use of PS as an alternative to the classical techniques used in beta emitter detection (Cerenkov and LS) is first discussed through their comparative application to widely determined beta emitters like $^{90}$Sr (0.546 MeV) and $^{90}$Y (2.27 MeV) in secular equilibrium. To this end, we quantified the activity of these isotopes in several water matrices using two procedures for quenching correction (SCR, T-SiE). The performance of the PS technique is also evaluated for the determination of a low-energy beta emitter ($^3$H), a beta-gamma emitter ($^{134}$Cs) and an alpha emitter ($^{238}$Pu) by single measurements in linear and logarithmic amplified detectors.

3.1. Comparative study of Cerenkov, liquid scintillation and plastic scintillation techniques for $^{90}$Sr/$^{90}$Y determination

This study compares the results for a series of test solutions quantified by Cerenkov, LS and PS using a linear amplified detector (Packard). For PS, an experimental approach to the limit of detection was also carried out. Finally, the influence of the type of detector, in relation to its amplification and background reduction, was also evaluated.

3.1.1. Spectrum shape

The spectra obtained in the measurement of the active and blank unquenched solutions by Cerenkov, LS and PS are shown in Fig. 1. For active solutions, the Cerenkov spectrum shape differed greatly from the LS and PS spectra. The peak obtained by the Cerenkov technique shows a single band located at low-energy, which corresponds to the low-energy photons produced by the Cerenkov phenomenon. In contrast, LS and PS peaks are located at higher energies and two overlapping bands due to the distinct energies of the beta particles emitted by $^{90}$Sr (lower part) and $^{90}$Y (higher part) are clearly distinguished. There was a slightly higher scintillation yield for PS than for LS under these experimental conditions.

The background spectra were similar for the three techniques and their influence on the quantification decreased as the active peaks were located in the higher energy region.

From the analysis of the spectral shape, it can be concluded that the LS and PS techniques show similar behaviour for $^{90}$Sr/$^{90}$Y detection, clearly different from that of the Cerenkov technique.

3.1.2. Detection efficiency and background

Relative detection efficiency values are based on the activity of $^{90}$Sr. The characteristics of the PS, LS and Cerenkov techniques in terms of detection efficiencies ($E$), background ($B$) and figure of merit (FM) in the full spectrum (0–4000) and in the OW are shown in Table 1. The results correspond to a calibration sample with a T-SiE value parameter similar to that of the test solution (M4, M3, EB and AS). Measurements were performed with a linear amplified detector (Packard Tri-Carb 2000).

For PS and LS, detection efficiency and background values were similar and different from those of the Cerenkov technique. In the full spectrum, the detection efficiencies of PS and LS were close to 200%, thus, practically all disintegrations were detected. For the Cerenkov technique, these values were ca. 70%, which corresponds with the Cerenkov phenomenon.
Fig. 1. Spectra of blank and 90Sr/90Y active unquenched solutions measured by Cerenkov, liquid scintillation and plastic scintillation techniques. The OWs were located at different regions of the spectrum owing to the different relative positions of the active and background peaks. In these OWs, the background decreased significantly for PS and LS, whereas detection efficiency did not decrease to the same extent. Therefore, FM was much higher in the LS and PS than in Cerenkov technique.

The relative errors produced when the 90Sr/90Y activities are quantified in five water matrices (AM, M3, M4, EB and AS) by LS, PS and the Cerenkov technique are shown in Table 2. Three independent replicates were studied for AS and single determinations for the other matrices.

Two calibration procedures were used, one based on the sample spectrum (SCR) and the other based on the external standard spectrum (T-SiE). No value of T-SiE was obtained when the scintillator was not present in the counting solution and so this procedure was not used for 3H.

Table 1

<table>
<thead>
<tr>
<th>Radionuclide</th>
<th>Detector</th>
<th>Technique</th>
<th>$E$ (%)</th>
<th>$B$ (cpm)</th>
<th>$E$ (%)</th>
<th>$B$ (optimum window)</th>
<th>Figure of merit</th>
<th>Limit of detection</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{90}$Sr/90Y</td>
<td>Linear Cerenkov</td>
<td>$60.1 \pm 1.1$</td>
<td>$7.4 \pm 0.2$</td>
<td>$60.1 \pm 1.0$</td>
<td>$725.8$</td>
<td>--</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Linear LS</td>
<td>$215.3 \pm 3.3$</td>
<td>$16.4 \pm 0.2$</td>
<td>$152.0 \pm 2.4$</td>
<td>$6718$</td>
<td>--</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Linear PS</td>
<td>$181.1 \pm 3.5$</td>
<td>$13.9 \pm 0.2$</td>
<td>$158.3 \pm 2.6$</td>
<td>$4297$</td>
<td>--</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Logarithmic PS</td>
<td>$191.0 \pm 3.1$</td>
<td>$1.7 \pm 0.1$</td>
<td>$179.3 \pm 2.9$</td>
<td>$24969$</td>
<td>--</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>$^3$H</td>
<td>Linear PS</td>
<td>$0.22 \pm 0.02$</td>
<td>$14.5 \pm 0.2$</td>
<td>$0.27 \pm 0.03$</td>
<td>$0.04$</td>
<td>635</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Logarithmic PS</td>
<td>$0.30 \pm 0.02$</td>
<td>$1.7 \pm 0.1$</td>
<td>$0.33 \pm 0.02$</td>
<td>$0.17$</td>
<td>297</td>
<td>--</td>
<td></td>
</tr>
<tr>
<td>$^{134}$Cs</td>
<td>Linear PS</td>
<td>$75.3 \pm 0.6$</td>
<td>$14.7 \pm 0.2$</td>
<td>$58.8 \pm 0.3$</td>
<td>$5.3 \pm 0.1$</td>
<td>667</td>
<td>4.3</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Logarithmic PS</td>
<td>$49.5 \pm 0.4$</td>
<td>$1.9 \pm 0.1$</td>
<td>$45.3 \pm 0.4$</td>
<td>$1.4 \pm 0.1$</td>
<td>1450</td>
<td>3.1</td>
<td></td>
</tr>
<tr>
<td>$^{238}$Pu</td>
<td>Linear PS</td>
<td>$41.9 \pm 1.5$</td>
<td>$14.7 \pm 0.2$</td>
<td>$24.4 \pm 0.8$</td>
<td>$2.4 \pm 0.1$</td>
<td>127</td>
<td>7.6</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Logarithmic PS</td>
<td>$46.9 \pm 1.2$</td>
<td>$1.5 \pm 0.1$</td>
<td>$28.5 \pm 0.8$</td>
<td>$0.33 \pm 0.03$</td>
<td>2429</td>
<td>2.4</td>
<td></td>
</tr>
</tbody>
</table>
Table 2

Relative error in $^{90}\text{Sr}/^{90}\text{Y}$ activities quantification in different water matrices (river (M3), purifying plant (M4), sea (AM), bottled (EB) and underground (AS)) by Cerenkov, liquid scintillation and plastic scintillation techniques

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Cerenkov T-SiE (%)</th>
<th>Cerenkov SCR (%)</th>
<th>Liquid scintillation T-SiE (%)</th>
<th>Liquid scintillation SCR (%)</th>
<th>Plastic scintillation T-SiE (%)</th>
<th>Plastic scintillation SCR (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>M3</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>0.73</td>
<td>0.15</td>
</tr>
<tr>
<td>M4</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>1.05</td>
<td>–1.0</td>
</tr>
<tr>
<td>AM</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>1.47</td>
<td>–1.10</td>
</tr>
<tr>
<td>EB</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>1.71</td>
<td>0.49</td>
</tr>
<tr>
<td>AS ($n=3$)</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>0.27</td>
<td>–0.60</td>
</tr>
</tbody>
</table>

Table 3

Quenching calibration function for different measurement techniques and quenching parameters

<table>
<thead>
<tr>
<th>Quenching parameter</th>
<th>Cerenkov</th>
<th>Liquid scintillation</th>
<th>Plastic scintillation</th>
</tr>
</thead>
<tbody>
<tr>
<td>T-SiE</td>
<td>$E = -0.0007x^2 + 0.595x + 31.321;$</td>
<td>$R^2 = 0.9959$</td>
<td>$E = -0.0002x^2 - 0.1083x + 139.37;$</td>
</tr>
<tr>
<td>SCR</td>
<td>$E = -0.626x^2 + 232.73x + 92.2;$</td>
<td>$R^2 = 0.9997$</td>
<td>$E = -272.37x^2 + 600.39x - 153.48;$</td>
</tr>
</tbody>
</table>

not applied in the Cerenkov technique. The functions obtained are shown in Table 3.

The relative accuracy obtained for the three techniques was similar, slightly higher for PS than for LS or Cerenkov, and always <4%. The higher values obtained with the Cerenkov technique may be due to the lower count rate for this technique, which may also account for the higher accuracy obtained with T-SiE than with the SCR calibration procedure.

The relative precision obtained for AS replicates was ca. 0.7% for LS and PS and higher for Cerenkov, which may be due to the lower count rate of this last technique.

From the results obtained it can be deduced that the performance of PS counting is comparable to that of the well-established LS technique for $^{90}\text{Sr}/^{90}\text{Y}$ determination in aqueous matrices.

3.1.3. Limit of detection

In addition to the activity level studied in the test set (300 dpm/g), two levels of lower activity, 23 and 2.3 dpm/g, were considered in the experimental approach to the limit of detection. The relative errors obtained in the quantification of each set are shown in Table 4. For each set, three spiked AS samples were measured.

These results reveal that both quenching calibration procedures, T-SiE and SCR, have similar behaviour. It is clear that the relative error increases when the activity level decreases. At 23 dpm/g, the results allow us to quantify the activity despite the poor precision achieved. At 2.3 dpm/g, semi-quantitative determination is still possible, but the uncertainty associated with the results is too high.

This behaviour places the experimental detection limit closer to 23 than to 2.3 dpm/g. This experimental detection limit for $^{90}\text{Sr}/^{90}\text{Y}$ using PS depends on the detector used, and it would be improved if an active guard detector (Quantulus) was used, as can be deduced from the background values quoted in Table 1. In any case, this experimental detection limit is close to the activity levels of environmental samples after the separation step and certainly...
allows us to quantify these isotopes in liquid effluents or in any aqueous solution coming from the separation process of these radionuclides in radioactive waste.

3.1.4. Comparative study of the linear and logarithmic detector

Once the capability of PS to quantify $^{90}$Sr/$^{90}$Y activity in a linear amplification detector (LN, Packard Tri-Carb 2000) was established, the detection efficiency and background in a logarithmic detector (LG, Quantulus EGG–Wallac) were determined. In addition to the logarithmic amplification, the main difference between these detectors was the background reduction system, pulse shape discrimination in the Packard Tri-Carb 2000 detector and pulse shape discrimination and active guard for Quantulus.

The spectra obtained with both detectors are shown in Figs. 2 and 3. The effect of the logarithmic amplification shifts and expands the lower energy range of the spectrum, where $^{90}$Sr is located, and shrinks higher energies, where the $^{90}$Y peak is located. As in the case of the linear detector, the contribution of each radionuclide is clearly distinguished. The figure also shows the background reduction by the active guard mode on the logarithmic detector.

The average of the results obtained for the test solutions is shown in Table 1. The values obtained with the LG detector clearly improve on the LN detector results. With regard to the LN detector values, the background in the full spectrum of the LG detector was significantly reduced, whereas detection efficiency slightly increased. The same behaviour was observed in the OW.

3.2. Other radionuclide performances

The capability of PS to determine radionuclides other than high-energy beta emitters was evaluated by quantifying a low-energy beta emitter ($^{3}$H, $E_{\text{max}}$ 0.0186 MeV), a gamma–beta emitter ($^{134}$Cs, $E_{\text{max}}$ 0.658 MeV) and an alpha emitter ($^{238}$Pu, 5.449–5.456 MeV). Measurements were made by both linear and logarithmic detectors. Three independent replicates were measured in the LN detector and single determinations were performed with the LG detector.

3.2.1. Tritium

The spectra obtained for $^{3}$H measurements in both detectors are, as expected, located in the first channels. Nevertheless, logarithmic amplification characteristics...
allow us to distinguish, in the same spectrum, the \(^{3}\)H and \(^{90}\)Sr/\(^{90}\)Y peaks, whereas in linear amplification, the \(^{3}\)H signal would remain totally hidden by the \(^{90}\)Sr/\(^{90}\)Y peak.

Detection efficiency was very low and similar in both cases (Table 1), owing to the low-energy of the beta particle emitted, whose energy was spent mostly in interactions with the medium before reaching the scintillator beads. Background values depend critically on the detector used, especially at the low-level energy range. LG improves the results obtained with the LN detector reducing the theoretical limit of detection to ca. 5 Bq/g.

3.2.2. \(^{134}\)Cs

\(^{134}\)Cs spectra obtained in both detectors were similar to the spectra obtained in \(^{90}\)Sr measurements owing to the similar energy of the beta particle emitted. Detection efficiency and background values for the beta-gamma emitter \(^{134}\)Cs are shown in Table 1. The efficiency value in the LG counter was unusually low, which may be due to the background reduction system of the LG detector. \(^{134}\)Cs disintegration is often detected simultaneously by the guard detector (gamma emission) and the main detector (beta particle). Both signals arrive in coincidence and the beta disintegration was mistaken for a cosmic background signal and cancelled.

The detection efficiency drawback was fully compensated by background reduction, which led to a FM and background better for the LG than for the LN detector.

Thus, the best behaviour of the plastic scintillator has been observed for high and medium energy beta emitters, whereas for low-energy radionuclides LSC is clearly better.

3.2.3. \(^{238}\)Pu

The spectra corresponding to \(^{238}\)Pu in the LG and LN detectors are similar and show a single peak due to the almost monoenergetic emission of this radionuclide.

Detection efficiency in the full spectrum and in the OW was very similar in both detectors (Table 1). The low efficiency value is due to the low penetration power of the alpha particles. Unless the isotope disintegration is close to the plastic scintillator, the energy is lost in interactions with the medium before reaching the beads.

The OWs were narrow in both detectors, and thus, the background values were lower than those obtained for the other isotopes. Again, LG detection registered
3.3. Cleaning procedure

A good reason to use the PS technique is that it does not produce mixed waste. After measurement, solid plastic scintillator beads can easily be separated from the initial aqueous solution by filtration. Moreover, PS beads and vials potentially show reversibility when an effective cleaning procedure brings the scintillator beads back to their initial state.

The first arrays using PS were performed in the linear detector with $^{90}$Sr/$^{90}$Y. The cleaning procedure applied initially consisted of bead and vial immersion in water bath for 12 h. Thereafter, the background was measured and compared with the reference range obtained in the previous measurements. This range was established as $13.920 \pm 0.215$ cpm and corresponds to the average $\pm$ two standard deviations of an experimental series of independent blanks prepared with new scintillator material.

After 20 measurements of $^{90}$Sr/$^{90}$Y followed by the cleaning procedure and blank preparation, 12 background values were included in the range of $\pm 1$ S.D., six in the range of $\pm 2$ S.D. and only three exceeded the $2 \sigma$ interval, showing memory effects. On applying a second, similar, cleaning process, the results for these three high blanks were within the reference interval. We would like to emphasise that the remaining activity in those that presented memory effects was <2% of the initial activity.

The plastic scintillator used in $^1$H measurements easily recovered the reference values following the cleaning protocol described earlier. The results obtained by applying this cleaning procedure were not so good for $^{134}$Cs and $^{238}$Pu. After a cleaning the remaining activity could achieve up to four standard deviations over the reference range. To remove all the activity of these isotopes and to avoid memory effects in $^{90}$Sr/$^{90}$Y, an extensive cleaning procedure was developed. The new cleaning procedure had to be compatible with the composition of the plastic scintillator beads to avoid their degradation. Therefore, organic solvents and concentrated acids were avoided, and the new procedure included four successive steps of water, 0.1 M HCl, ultrasonic and non-ionic detergent treatment.

This procedure improved the previous values. The remaining activity of $^{90}$Sr/$^{90}$Y, $^1$H and $^{238}$Pu was always removed in the first or the second washing attempt, whereas for $^{134}$Cs, up to four successive processes were needed in some cases. At any case, the counts registered never surpassed the reference value by more than 1 dpm.

4. Conclusions

According to the results obtained in this study, the PS technique can be used as an alternative to LS and the Cerenkov technique in beta emitter determination. Detection efficiency, background and relative errors in sample quantification achieved by using PS are similar to those obtained by LS and better than those provided by Cerenkov. Moreover, PS technique does not produce mixed waste from to the measurement process, and the material can be reused in many cases after cleaning. Nowadays, the main drawback to introducing PS in routine measurements is its high cost, which may decrease if the demand rises. This study also shows the capability of plastic scintillators to quantify alpha- and beta-gamma emitters with similar performance to LS counting.

Acknowledgements

The authors thank the CICYT for financial support (Grant AMB 99-0430) and Aguas de Barcelona for its collaboration in water sampling.

References


