## PLASTIC SCINTILLATORS: A POWERFUL TOOL TO REDUCE MIXED WASTE.

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**ABSTRACT**

Wastes containing radioactive and organic compounds (mixed wastes) are difficult to dispose because of the regulations established for nuclear and hazardous wastes. Mixed wastes originate mainly in the emulsions generated in beta emitter determinations by Liquid Scintillation techniques. The use of plastic scintillators instead of liquid cocktails may facilitate the segregation, after measurement, of sample and scintillator without introducing additional wastes in the measurement step.

In this study, we compare the capability of Plastic Scintillation (PS) versus Liquid Scintillation (LS) and Cerenkov (C) techniques to determine beta emitters in routine measurements. Results obtained show that high and medium energy beta emitters (Sr-90/Y-90 and C-14) can be quantified in aqueous samples by using PS with similar relative errors (< 5%) as those obtained by LS or C, for any activity level considered. For low energy emitters (H-3), best results using PS are achieved for medium activity levels. Additionally, measurements performed in solutions including alpha (Pu-238) and beta-gamma (Cs-134) emitters confirm the capability of PS to extent the application of this technique to the determination of these types of isotopes.

**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 because of the regulations established for nuclear and hazardous wastes [1, 2]. Although their activity can be low, the large amount of mixed wastes 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 techniques in biomedical [4] and environmental studies [5]. In these emulsions, the waste results from mixing the organic liquid scintillation cocktails and aqueous radioactive samples [6, 7]. The use of plastic scintillators instead of liquid cocktails may facilitate the segregation, after the measurement, of sample and scintillator without introducing additional wastes in the measurement step

Plastic scintillation (PS), which was first used in 1960 [8, 9], has always been considered a very versatile technique, due to the large number of polymers and shapes that could be synthesized [10]. However, the poor results obtained in its first analytical applications and its good response as a large surface area cosmic particle detector prevented it being widely used for determining beta emitters activity in routine measurements. New plastic scintillators not only overcome some of these drawbacks, but also introduce new possibilities for beta radionuclide quantification. The development of plastic scintillator beads and fibres [11] has led to the design of analytical

technology able to determine beta emitter activity by continuous flow [12], remote [13, 14] and routine measurements [15, 16], all of which achieve high detection efficiencies for high-energy beta emitters.

The aim of our work is to present Plastic Scintillators as a real alternative to the classical techniques to determine beta emitters: Liquid Scintillation and Cerenkov [17]. The performances of the three methods were compared for the determination of a beta emitter Sr-90/Y-90 in secular equilibrium, a low-energy beta emitter, H-3, an alpha emitter, Pu-238, and a beta + gamma emitter, Cs-134.

The capability of plastic scintillation beads and polyethylene vials as a routine technique to determine the activity of three of the most important beta emitters, (Sr-90/Y-90 (high energy), C-14 (medium energy) and H-3 (low energy)), in low level aqueous spiked samples was evaluated.

Finally, in order to reuse the beads, we establish a cleaning procedure for plastic scintillation beads.

This article summarises the content included in the references [18, 19, 20 and 21].

## EXPERIMENTAL

### Reagents and solutions

All regents used were of analytical or scintillation grade.

Sr-90/Y-90, C-14, H-3, Cs-134 and Pu-238 active stocks solutions were used for the preparation of active solutions. Sr-90 and Y-90 are in secular equilibrium, although nominal activity was related to the Sr-90 activity. Active solution was prepared by diluting a weighed amount of standard solution in the corresponding carrier solution.

Underground water from the proximity of Ascó nuclear plant (NE Spain) (AS) was used for the preparation of the test solutions. 0.1 g/l Bromocresol green in water were used as colour quenchers.

For Liquid Scintillation measurements, Opthiphase-Hisafe® (from EGG-WALLAC) was used as liquid scintillation cocktail. For Plastic Scintillation measurements, solid plastic scintillator beads based on polyvinyltoluene were used as scintillator reagent. The diameter of the beads was between 250 and 500 μm. Beads were supplied by Bicron ®(BC-400).

For Cerenkov, LSC and PS measurements, 7ml vials (from Packard Instruments Co.) were used. For Plastic Scintillation, vials of plastic scintillator BC-408 (from Bicron®) were also used. The shape of these vials was equivalent to the 7 ml polyethylene vials.

Count-Off[TM] was supplied by NEN[TM] Life Science Products

### Apparatus

A Tri-Carb 2000 CA/LL liquid scintillation detector (Packard) with linear amplification, a multichannel analyzer of 4096 channels (0-2000 Kev) and background reduction based on pulse-shape discrimination and a Quantulus liquid scintillation detector (EG&G Wallac) with logarithmic amplification, a multichannel analyzer of 4096 channels distributed in four segments of 1024 channels, alpha/beta discrimination and background reduction by active guard were used.

### Measurement solution preparation

For Cerenkov measurements the volume of the counting solutions was 3ml in all cases. For Liquid Scintillation measurements composition of the counting solutions includes 0.8ml of aqueous solution and 2.2ml of scintillation cocktail. In case of PS, 0.425g of beads and 0.2 ml of aqueous solution was added when the PS vials were used, whereas 1.8g of beads and 1 ml of aqueous solution were added to polyethylene vials.

Aqueous calibration solutions were prepared by adding the corresponding volume of active solution, distilled water, and quenching agent (Bromocresol green 0.1g/l). In test solutions, distilled water fraction was replaced by the underground water AS. Blank solutions were done substituting the active solution by distilled water.

The amount of each fraction was always determined by weighing. Once prepared, the vials were shaken and left to stand in the dark for 2h before measurement.

### Measurement step

For measurements done in a Packard Tricarb-2000 detector, the influence of the low level option was first evaluated. The optimal conditions found for the different techniques were: Cerenkov, Low Level (LL) on, Liquid Scintillation and Plastic Scintillation, LL off. In all cases, measurement time was five successive periods of one hour.

### Quenching correction and Data treatment

The spectra collected for each solution were smoothed using a Savitzky-Golay algorithm and then averaged. Net spectra were obtained by subtracting to each active sample its equivalent blank solution. Quenching correction curves were obtained by fitting optimum window (OW) detection efficiencies to their respective values of the quenching parameters (SQP(E) and SCR) for each set of calibration solutions. The detection efficiency of each test solution was calculated as the ratio between the count rate registered in the OW of the net spectra and the total activity of the test solution. Optimum windows were established for each radionuclide, using the unquenched calibration solutions. OW corresponds to the range of the spectra with best figure of merit (FM $= E^2/B$, where E is the efficiency and B the background).

For each solution, the SCR (Sample Channel Ratio) 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. The SQP(E) parameter (standard external quenching parameter) was calculated by the logarithmic detector and corresponds to the end-point of the external gamma source (Ra-226) spectrum.

For test solutions, activity was calculated by using the integrated net signal in the optimum window and its detection efficiency. This efficiency was obtained by interpolating the quenching parameter value in the corresponding quenching correction function.

Theoretical detection limit was calculated using the expression recommended by the Spanish nuclear authorities for liquid effluents[22]: $DL=2*s(B)/(W*E)$, where $s(B)$ is the standard deviation of a blank with a quenching parameter value similar to that of the test solution value, w is 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.

**Cleaning procedure**

Vials and beads of Plastic Scintillator were reused. To remove the remaining activity, a initial specific cleaning procedure was applied for each isotope.  This procedure was applied until an optimized procedure, described later, was finally adopted

For Cs-134 and Pu-238 measurements, the protocol followed was:
1. - rinse the vials with HCl 0.1 M.
2. - Shake the vials in a 250 ml-polyethylene bottle filled up with 0.1M HCl for 15 minutes.
3. - rinse the vial with distilled water
4. - apply ultrasonic bath to the vials in a 250 ml-polyethylene bottle filled up with distilled water for 15 min.
5. - As step two, but using non-ionic soap solution (COUNT-OFF®) for 12 hours.
6. - As step 5, but using distilled water.
7. - dry the vials using a paper filter.

The protocol for 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 40º.

For Sr-90/Y-90 and H-3 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.

**SAFETY CONSIDERATIONS**

All the experimental work was done according to the regulations of the Spanish Nuclear Authorities (Consejo Seguridad Nacional) and the statutes of the University of Barcelona.

**RESULTS AND DISCUSSION**

**Comparative study of Cerenkov, Liquid Scintillation and Plastic Scintillation techniques for beta, alpha and gamma emitters determination**

The first part of the work compares the results of a series of Sr-90/Y-90 test solutions quantified by Cerenkov (C), Liquid Scintillation (LS) and Plastic Scintillation (PS) using a linear amplified detector (Packard).

The spectra obtained in the measurement of the active and blank unquenched solutions are shown in Figure 1. For active solutions, Cerenkov spectrum shape greatly differed from Liquid and Plastic Scintillation spectra. The peak obtained in Cerenkov shows a single band located at low energy region, which corresponds to the low energy photons produced by the Cerenkov phenomena. In contrast, Liquid and Plastic Scintillation peaks are located at higher energies and two overlapped bands, due to the distinct energy of the beta particles emitted by Sr-90 (lower part) and Y-90 (higher part), are clearly distinguished.

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

From the analysis of the spectra shape, it can be concluded that Liquid and Plastic Scintillation techniques show a similar behaviour for Sr-90/Y-90 detection, clearly different from that of the Cerenkov technique.
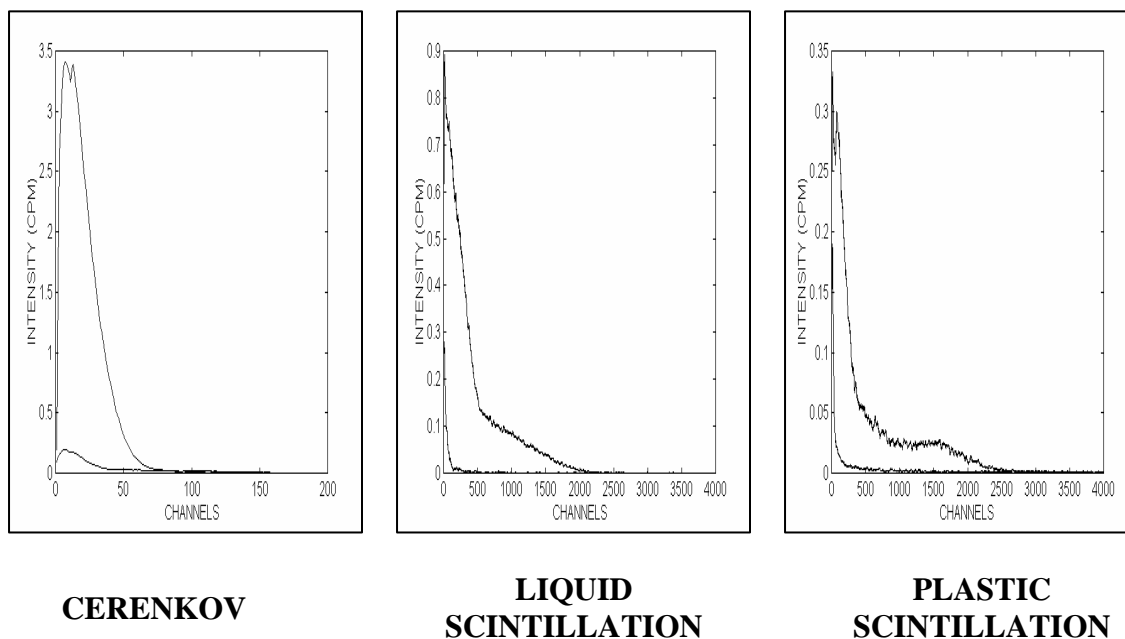


**CERENKOV**  **LIQUID SCINTILLATION**  **PLASTIC SCINTILLATION**

**Fig. 1.** Spectra of blank and Sr-90/Y-90 active unquenched solutions measured by Cerenkov, Liquid Scintillation and Plastic Scintillation techniques.

Detection efficiency and background

Relative detection efficiency values are based on the activity of the Sr-90 isotope. The characteristics of PS, LS and C in terms of detection Efficiencies (E) and Background (B) in the optimum window are shown in Table 1. Results correspond to a calibration sample with a quenching parameter value similar to that of the test solution (underground water AS).

For Plastic and Liquid Scintillation, detection efficiency and background values were similar and different from those of Cerenkov. In the full spectrum, the detection efficiencies of PS and LS were close to 200%, thus practically all disintegrations were detected. For Cerenkov, these values were around 70%, which corresponds to Cerenkov phenomena.

The optimum windows (OW) were located at different regions of the spectrum owing to the different relative positions of the active and the background peaks. In these OW, background decreased significantly for PS and LS, whereas detection efficiency did not decrease in the same extension. Therefore, FM was much higher in LS and PS than in Cerenkov.

The relative error produced when the Sr-90/Y-90 activities are quantified in AS by LS, PS and C is shown in Table 1. Three independent replicates were done. The relative accuracy obtained for the three techniques was similar, slightly higher for PS than for LS or C, and always lower than 4%. The higher values obtained with Cerenkov may be due to the lower count rate detected for this technique. Relative precision obtained was around 0.7% for LS and PS and higher for C, which may be due also to the lower count rate of this technique.

From the results obtained it can be deduced that the performance of Plastic Scintillation is comparable to that of the well-established Liquid Scintillation technique for Sr-90/Y-90 determination in aqueous matrices.

**Table I.** Background, Detection Efficiency of Sr-90/Y-90, H-3, Cs-134 and Pu-238 by Cerenkov, Liquid Scintillation and Plastic Scintillation in the Packard Detector

| Radionuclide | Technique | Optimum Window (channels) | EFF (%) (Optimum Window) | Background (cpm)(Optimum Window) | Relative Errors (%) |
|---|---|---|---|---|---|
| Sr-90/Y-90 | CERENKOV | 2-53 | 60.1 ± 1.0 | 5.0 ± 0.1 | -3.59 ± 2.58 |
| Sr-90/Y-90 | LS | 122-1649 | 152.0 ± 2.4 | 3.4 ± 0.1 | -1.64 ± 0.69 |
| Sr-90/Y-90 | PS | 67-2180 | 158.3 ± 2.6 | 5.8 ± 0.1 | -0.60 ± 0.50 |
| H-3 | PS | 1-14 | 0.27 ± 0.03 | 2.0 ± 0.1 | --- |
| Cs-134 | PS | 49-840 | 58.8 ± 0.3 | 5.3 ± 0.1 | --- |
| Pu-238 | PS | 150-571 | 24.4 ± 0.8 | 2.4 ± 0.1 | --- |

The capability of PS to determine radionuclides other than high-energy beta emitters was evaluated by quantifying a low energy beta emitter (H-3, Emax 0.0186 Mev), a gamma-beta emitter (Cs-134, Emax 0.658 Mev) and an alpha emitter (Pu-238, 5.449-5.456 Mev). Three independent replicates were measured.

Tritium

The spectra obtained for H-3 measurements is, as expected, located at the first channels. That makes that the H-3 signal would remain totally hidden by the Sr-90/Y-90 peak. Detection efficiency was very low 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.

Caesium-134

A Cs-134 spectrum obtained was similar to the spectra obtained in Sr-90 measurements owing to the similar energy of the beta particle emitted. The same happens with the detection efficiency and background values. Thus it can be concluded that the best behaviour of plastic scintillator is observed for high and medium energy beta emitters, whereas for low energy radionuclides LSC is clearly better.

Plutonium-238

The spectra corresponding to Pu-238 show a single peak due to the almost monoenergetic emission of this radionuclide. For that reason the optimum window is very narrow and as consequence the background value was lower than those obtained for the other isotopes. Low detection efficiency value was 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.

**Use of Plastic Scintillation beads and polyethylene vials as a routine technique**

Once it is clear that plastic scintillation beads are suitable for beta emitter determination the main drawback to use it as a routine technique is nowadays its high cost. This can be reduced by the use of polyethylene vials and the reuse of the beads in multiple analyse. Of course, the increase of the PS beads production as consequence of the adoption of this approach will also reduce drastically the cost.

The ability of the plastic scintillation beads and polyethylene vials as a routine technique to determine the activity of Sr-90/Y-90 (Emax=0.55 Mev/ Emax=2.2 Mev), C-14 (Emax=156 Kev) and H-3 (Emax=18.6 Kev) was assessed by comparing the activity calculated from the measurement of the test samples with the activity spiked into the sample. Optimal detector measurement conditions for the logarithmic detector and colour quenching calibration functions were established for all the isotopes prior to test sample quantification.

Sr-90/Y-90

Optimum counting conditions were established by comparing the detection efficiency and background values obtained when measuring the unquenched calibration solution. The best results for Sr-90/Y-90 were obtained with low coincident bias and the C-14 MCA configuration. Other options gave a slightly worse detection efficiency-background ratio because Sr-90/Y-90, as high energy beta emitters, are not affected by the high coincident bias option, which cancels part of the active and background spectra on the first channels, nor by the H-3 MCA configuration option, which introduces more restrictive conversion rules into the signal analysis.

The spectra obtained for the blank and active Sr-90/Y-90 unquenched calibration solutions are shown in Figure 2. The background signal is very low and is concentrated in the lower part of the spectrum; therefore, its influence on the Sr-90/Y-90 spectra, mainly located over two hundred channel, is not important. Active spectra clearly show two peaks, which correspond to Sr-90 and Y-90

Figures derived from these spectra can be seen in Table 2. The high values obtained for detection efficiency in the full spectrum (near 90%) indicate that almost all beta disintegrations reach the scintillator surface. This means that the bead size is adequate for high energy beta emitter detection and that the vials have no significant influence on the detection efficiency. Previous work done by using plastic scintillation vials (BC-400), which have the same shape than the polyethylene vials used in this study, yields similar values for the analytical parameters.

The optimum window obtained is very broad and covers more than half the total number of channels. However, the different location of active and background spectra permits to achieve extraordinarily detection efficiency/background ratios at the optimum window.
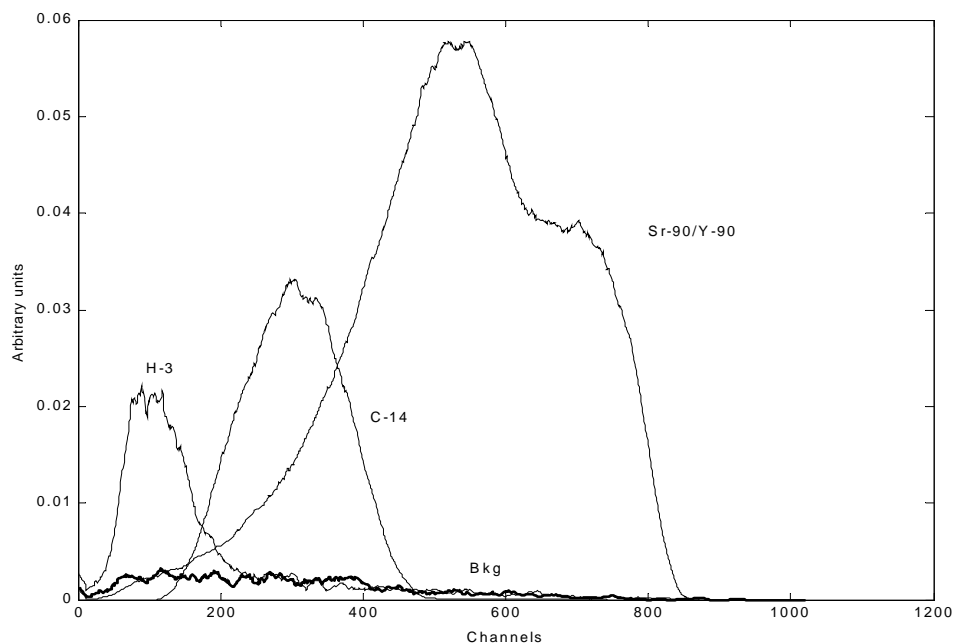


**Fig. 2.** Sr-90/Y-90, C-14, H-3 active and background spectra

Background and efficiency obtained in the optimum window will permit to achieve a detection limit of 6.8 x 10-4 Bq/ml. This value is better than the limit quoted by the Spanish nuclear authorities for liquid effluents monitoring (8.5 x 10-4 Bq/ml).

Two efficiency calibration procedures, one based on the sample spectrum (SCR) and the other on the external gamma source spectrum (SQP(E)), were applied in order to quantify the set of spiked water samples.

Mean values for relative errors obtained in quantifying single spiked samples and the three AS replicates are shown in Table 4. High accuracy is observed in all cases, as both absolute and relative errors obtained are lower than 1cpm and 3%. The precision obtained for AS determinations corresponds to the theoretical uncertainty associated to the activity level spiked.

On the basis of these values it can be concluded that plastic scintillation beads and polyethylene vials may be used as an alternative to classical methods for low level Sr-90/Y-90 determination in routine aqueous samples.

**Table 2.** Measurement Conditions, Detection Efficiency, Background and Mean Relative Errors in the Optimum Window for Sr-90/Y-90, C-14 and H-3 Measurements in the Quantulus Detector.

|  | Measure time(h) | Activity (dpm/g) | Optimum Window | Efficiency (%) (O.W.) | Bkg. (cpm) (O.W.) | Relative Errors (%) | |
|---|---|---|---|---|---|---|---|
|  |  |  |  |  |  | SQE | SCR |
| Sr-90/Y-90 | 5 | 100 | 396-849 | 124.61±3.76 | 0.19±0.03 | 1.38±3.47 | -0.77±3.26 |
| C-14 | 8 | 300 | 175-405 | 15.97±0.49 | 0.23±0.03 | 0.64±0.51 | -1.70±0.51 |
| H-3 | 24 | 2730 | 44-189 | 0.13±0.04 | 0.37±0.02 | 6.14±4.61 | 12.02±14.16 |

C-14

Optimum detector measurement conditions were established as C-14 MCA configuration and high coincident bias. As with Sr-90/Y-90, the detection efficiency-background ratio did not vary with the MCA configuration and the final option was chosen by considering the detector recommendations for high-medium energy beta emitters. On the other hand, high coincident bias greatly influenced signal registration by cancelling a significant number of low energy signals. This cancellation mainly affects the background, whose signals are low energy, and, to a lesser extent, the active sample, whose signals are more energetic. The final result was an increase in the detection efficiency-background ratio and, therefore, an improvement in measurement conditions.

Background and C-14 spectra of a calibration solution are plotted in Figure 2 and confirm the above mentioned hypothesis.

With regard to the Sr-90/Y-90 peak, C-14 is narrower and is located at lower energies. This position implies a decrease in detection efficiency, although it would help in the resolution of C-14 and Sr-90/Y-90 mixtures.

Detection efficiency, background and optimum window position figures are shown in Table 2. Values in the optimum and the full spectrum are similar due to the low background values obtained and to the fact that active and blank spectra coincide. It is worth noting the important decrease in detection efficiency for C-14, 18%, with regard to the value obtained for Sr-90/Y-90, 90%. This behaviour must be related to a decrease in the radiation energy of the isotope measured. As beta energy decreases, the number of particles unable to reach the scintillator beads increases. This phenomenon constitutes a limitation of PS applications. Nevertheless, the

efficiency stated will still enable plastic scintillation to be used for C-14 quantification in low activity aqueous samples.

Mean values obtained for the test set are shown in Table 2. Absolute and relative errors were very low; less than 6 cpm and 4%, and experimental uncertainty (relative error ± two standard deviations) was below 5% for any sample. This value coincides with the theoretical uncertainty associated with the measurement procedure and demonstrates the robustness of the technique.

Contrary to Sr-90/Y-90 results, the theoretical detection limit achieved (0.57x10-2 Bq/ml) exceeds that recommended by the Spanish nuclear authorities recommendations for liquid effluents (0.37x10-2 Bq/ml), although results can clearly be improved by increasing counting time and sample volume.

H-3

Tritium is a good example of a low energy beta emitter (18.6 KeV), and its study will permit the evaluation of the ability of the plastic scintillation and polyethylene vials technique to quantify these radionuclides. The best measurement conditions were low coincident bias and the H-3 MCA configuration. For this isotope, the use of high coincident bias was not possible because the coincident device could not distinguish between the background signal and tritium disintegration signals and cancels both. Consequently, very few counts were registered using this option. The H-3 MC configuration was chosen following the detector recommendations, although no difference was observed with respect to the C-14 option.

The background and active spectra obtained are plotted in Figure 2. The low energy location of both the tritium and the background spectra is responsible of the poor results obtained when the high coincident bias option was used. The location of the active tritium spectra in the first channels is due to its low energy and they can be seen even in LSC. In terms of the other isotopes measured, there was no significant overlapping between the peaks in either the total or the optimum window. The position of the H-3 peak would help to resolve complex samples, including H-3, C-14 and Sr-90/Y-90.

The value obtained for the detection parameters in H-3 determination with PS and polyethylene vials are shown in Table 2. In LS, the H-3 peak is also located in the low energy range of the spectrum, although the detection efficiency is not as low as in the case of PS and polyethylene vials. This behaviour is again related to the energy of the beta particle emitted by the radionuclide. For tritium, the scintillator and emitter are close enough in LS to produce 60% detection efficiency, whereas in PS the distance is too great and radiation energy is dissipated in interactions with the medium before reaching the plastic beads. Reduction of the beads-radionuclide distance, in order to increase the detection efficiency, could be achieved by immobilising selective compounds on the beads surface, as in SPA technology, or by reducing the bead diameter. The first option depends on the chemical behaviour of the element and cannot always be applied. Therefore, reducing the bead diameter will be the best approach when low energy beta emitters must be measured.

Slightly worst correlation, compared with those of Sr-90 and C-14, was found between detection efficiency and quenching parameters for the two cases studied, SCR and SQP(E). This behaviour

was related to the high uncertainty of the measurement, which is associated to the low count rate registered. Better correlation was obtained for the SQP(E) calibration procedure due to high activity of the external standard source.

This fact is also responsible of the high relative errors obtained in sample quantification when SCR calibration procedure is applied. This high experimental uncertainty values agree with the theoretical uncertainty. Better results are obtained by using the SQP(E) procedure. Relative errors achieved in this case were lower than 10%.

On the other hand, the detection limit achieved for this radionuclide is 1.4 Bq/ml (for 1ml of sample and 5 hours of measurement), slightly higher than the value recommended by the Spanish authorities [22] for liquid effluents (0.37 Bq/ml). Nevertheless, this limit could be achieved whenever the counting time were increased to 48 hours (theoretical limit 0.30 Bq/ml).

On the basis of these results, the PS and polyethylene vials technique can only be recommended for tritium routine activity determination in medium level activity samples and when an external standard quenching parameter calibration procedure was used.

**Reusability of the beads**

In order to reduce the cost of the analysis and the level of organic waste produced, we have developed a more complete cleaning procedure that will allow the return of the beads to its initial state and reuse it in new analysis.

Five cleaning solution, Water, HCl 0.1M, Count-Off solution, EDTA 0.01M (pH=3), and Citric acid 0.1M (pH=3), were evaluated to clean the beads that previously have been contaminated during 24 with an active solution (DPM). After contamination, beads are segregated from the radioactive solution and a cleaning step consisted of immersing 1.2g of beads in 100ml of the corresponding cleaning solution is applied. Beads and cleaning solution were placed in a glass bottle and shaken in an end-over-end shaker for 24 hours. Afterwards, beads were filtered and dried at 40ºC for 2 hours in an oven and measured to evaluate if the activity is removed.

Values obtained are compared with those achieved by using completely new beads ("ref-new" beads) and those cleaned by the previous cleaning procedure described in the experiment section ("ref-used" beads). Measurement was done by triplicate and solutions were prepared by adding 0.35g of plastic scintillator beads and 0.2g of the corresponding carrier solution to the polyethylene vials.

First experiment OPTI1 consists on a unique cleaning step. Results are shown in table 3. Count rate (cpm) and relative memory values were obtained for each isotope in its corresponding optimum window (O.W.). Relative memory was calculated as the ratio between the remaining activities on beads, cpm/g, with regard to the activity added in the contamination procedure, expressed in dpm/g:

$$R.M.(\%) = \frac{(A - B)/Eff}{Act} * 100$$

where,

R.M.= Relative memory

A= cpm/g of the beads cleaned by a specific cleaning solution
B= cpm/g of the "ref-used" beads
Act= dpm/g of the contaminated beads
Eff= Detection efficiency

**Table 3.** Cleaning and Test Optimization Experiments (Results given are count rate (cpm/ml) and RM values (in parenthesis))

| | Opti1 | Opti2 | Opti3 | TestS |
|---|---|---|---|---|
| **"REF-NEW" BEADS** | 0.20 ± 0.01 | 0.203 ± 0.007 | 0.203 ± 0.007 | 0.203 ± 0.007 |
| **"REF-USED"BEADS** | 0.29 ± 0.05 | 0.295 ± 0.196 | 0.326 ± 0.085 | 0.307 ± 0.091 |
| **CITRIC A. 0.1M** | -- | -- | **0.36±0.11 (0.02%)** | **0.39 ± 0.04 (0.05%)** |
| **CITRIC A. 0.1M** | -- | **0.34±0.03 (0.03%)** | **0.34±0.11 (0.01%)** | **0.36 ± 0.02 (0.03%)** |
| **CITRIC A. 0.1M** | **0.28±0.03 (0.01%)** | **0.24±0.04 (-0.01%)** | **0.22±0.03 (-0.05%)** | **0.26 ± 0.03 (-0.03%)** |
| **WATER** | 0.65±0.31 (0.21%) | 0.35±0.05 (0.03%) | 0.21±0.08 (-0.05%) | -- |
| **HCL 0.1M** | 0.32± 0.04 (0.02%) | 0.32±0.04 (0.02%) | 0.29±0.04 (-0.04%) | -- |
| **COUNT-OFF** | 0.31± 0.02 (0.01%) | 0.29±0.04 (-0.01%) | 0.30±0.04 (-0.01%) | -- |
| **EDTA 0.1M** | 0.39±0.06 (0.06%) | 0.27±0.04 (-0.01%) | 0.223±0.07 (-0.04%) | -- |

The lowest cpm value was obtained for the citric acid solution, however the use of a citric acid 0.1M (pH=3) solution can bring the beads to the "ref-used" level but not to the "ref-new" level so an additional cleaning step was considered. It must be pointed out that the main part of the activity is removed and no significant Sr-90/Y-90 adsorption happens.

In the Opti2 experiment, contaminated beads are first cleaned by a citric acid solution and then cleaned again separately with the five studied solutions. The values achieved (Table 3) after the two cleaning steps are very close to the initial values of "ref-used" beads. However at any case "ref-new" is achieved. Again the citric acid solution produces the best results, and consequently, it is selected for next experiment as the cleaning solution for the first and second steps.

Table 3 shows the Opti3 results. In this case contaminated beads were cleaned twice by a citric acid solution and then an additional step was done with the five proposed solutions. Citric acid 0.1M (pH=3) and water are able to bring the beads to almost the "ref-new" level.

According to these results the proposed cleaning procedure for plastic scintillation beads consists of three successive baths of 1.2 g of beads in 100ml of 0.1M (pH=3) citric acid solution for 24 h.

In order to establish the capability of the optimum protocol to remove the residual activity of Sr-90/Y-90 and the reproducibility of the cleaning procedure proposed, Sr-90/Y-90 contaminated beads were divided into five parts and the optimum cleaning protocol was applied to each group separately (TestS). After the three cleaning steps proposed, the count rate value achieved is lower than the obtained with the "ref-used" beads and quite close to that of the "ref-new" beads. On the other hand, the relative standard deviation of these five replicates is approximately the same than the RSD obtained when only measurement dispersion was considered (optimisation experiments). Thus, no more uncertainty is introduced during the cleaning process.

## CONCLUSIONS

According to the results obtained in this study, Plastic Scintillation technique can be used as an alternative to Liquid Scintillation and Cerenkov in beta emitter determination. Detection efficiency, background and relative errors in high and medium beta emitters quantification by using Plastic Scintillation are similar to those obtained by Liquid Scintillation and better than those provided by Cerenkov.

Moreover, Plastic Scintillation technique does not produce mixed waste reducing the danger and the cost of the waste treatment.

The use of polyethylene vials, rather than the plastic scintillation ones as container of the scintillation beads, does not significantly affect the detection efficiency values obtained and provides a cheap and easily option for routine determinations.

A cleaning procedure, capable of eliminating any memory effect and to bring the plastic beads to a stable value, is proposed to clean plastic scintillator beads. This protocol consists of three consecutive steps in which 1.2g of beads are immersed in 100ml of a citric acid 0.1M (pH=3) solution and shaken in a end-over-end shaker for 24 h. The reusability of plastic scintillator beads means a great cost reduction in the measurement procedure and an absolute reduction in the amounts of mixed waste produced.

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