PERFORMANCE EVALUATION OF LSC METHODS FOR SIMULTANEOUS DETERMINATION OF GROSS ALPHA-BETA ACTIVITY IN SPIKED WATER SAMPLES

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Abstract

Gross alpha/beta activity determination is utilized as a very popular Liquid Scintillation Counting (LSC) technique widely used in routine monitoring of drinking, surface and waste waters or in radiological emergency situations as a rapid screening method. In the present paper there are presented comparatively the performances of two methods for determining gross alpha-beta activity using LSC. One is based on the possibility of spectral deconvolution which represents a good solution when the liquid scintillation analyzer does not have the alpha-beta discrimination capability, and the other is based on the establishment of optimal pulse decay discrimination (PDD), depending on the extinction level of the sample. Conducting experimental tests provided an opportunity to compare the validity, reliability and limitations of both methods.

Keywords: *Gross alpha, Gross beta, Spectrum deconvolution, LSC*

Introduction

Determination of gross alpha activity using the Liquid Scintillation Counting (LSC) method is widely used both in the routine monitoring of drinking, surface and waste water, as well as in radiological emergency situations as a rapid screening method. One of the main procedures found in the scientific literature for determining gross alpha-beta activity is based on evaporation or co-precipitation in a thin source deposit prior to measurement by gas flow proportional counting. However, this method may not be appropriated in the case when the amount of residue resulting from the evaporation of the sample is too high, requiring additional tests to determine the self-absorption correction factor, or in case of an emergency because of the time necessary for the sample preparation, the extra detection equipment needed in case of analysing several samples at the same time, and the possible cross contamination of the samples. In these cases, the method of determining gross alpha-beta activity using LSC is preferred both for the simplicity of sample preparation and for the high counting efficiency [1].

Whether it is about determination of total activity or spectrometric measurements, the LSC method involves mixing a radioactive sample with a scintillation cocktail, which can convert the nuclear decay energy into photons of light that are detected and converted into electrical pulses by photomultiplier tubes (PMTs). The quenching phenomenon occurs during the transfer of energy from solvent to the scintillator. There are different factors that hinder the energy transfer between the emitted particle and photons detected by the PMT, leading to a decrease in detection efficiency. There are studies which have shown that quenching also affects the degree of α/β misclassification, which is a consequence of different manners of its influence on early and delayed components of the light pulse [2, 3].

In the present paper, the performances of two methods for determining gross alpha-beta activity using LSC method are comparatively presented.

The first method is based on the establishment of optimal pulse decay discrimination (PDD), depending on the extinction level of the sample. For this purpose, there have been performed experimental tests to determine the optimal PDD using calibration standards containing alpha and beta emitters (^{241}Am and ^{90}Sr/Y).

The second method is based on the possibility of spectral deconvolution which represents a good solution in case when the liquid scintillation analyzer does not have the alpha-beta discrimination capability.

Materials and methods

Equipment

The experimental tests have been performed using a liquid scintillation analyzer, model PerkinElmer TriCarb 4910 TR and radioactive standard solutions containing different alpha and beta emitters (^{241}Am, ^{90}Sr/Y).

Selection of the quenching agent

The most often used quenching agents that are available in the laboratory are: carbon tetrachloride (CCl4), acetone (CH3CH3CO), chloroform (CHCl3), and nitromethane (CH3NO2) [4]. The performance of two selected quench agents, acetone and carbon tetrachloride, have been investigated by adding gradually known masses of them in two different samples consisting in 15 ml of scintillation cocktail (Ultima Gold AB).

After each addition, the value of tSIE parameter for the two samples was determined in order to assess the quenching levels the agents induce through the transfer of the kinetic energy of the decay particles to the scintillation cocktail solvent. For these tests, polyethylene vials with a volume of 20 ml have been used.

The quench level induced by adding increasing amounts of the two quenching agents to the sample could be observed in Fig. 1.

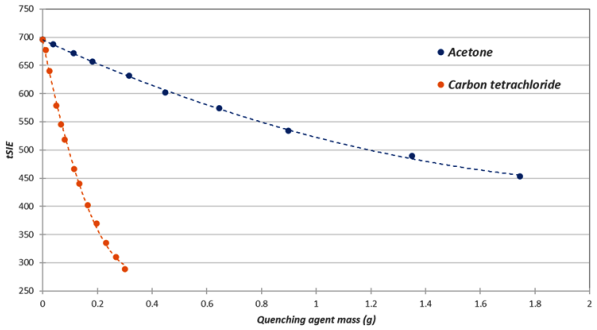


Fig. 1. Quench level induced by the two quenching agents

Method for simultaneous determination of gross α-β activity using the optimal PD

Spillover was used to quantify the PDD of the samples. Alpha spillover into the beta channel, X_α, is calculated as the ratio of number of alpha events that are classified as beta events to the total number of events, and is quantified for a pure alpha emitter. Beta spillover into the alpha channel, X_β, is similarly quantified with a pure beta emitter. The working spillover is defined as the point where $X_\alpha = X_\beta$. The working pulse decay discriminator (PDD) setting is defined as the PDD value for the working spillover [5].

Determination of optimum value of PDD requires pure alpha and beta standard. In order to obtain the most accurate results, the standards must be as near identical as possible to the unknown samples in terms of chemical composition, volume, vial type, etc.

Following the measurement of the samples, the count rate observed in α and β -MCA is a function of both alpha and beta disintegrations plus their interference in β and α -MCA respectively. The following notations are made in order to be able to calculate gross alpha and beta activities:
A_T - true count rate due to alpha decays in the sample
B_T - true count rate due to beta decays in the sample
A_{0S} - count rate observed in α -MCA when measuring the sample
B_{0S} - counting rate observed in β -MCA when measuring the sample
A_B - counting rate observed in alpha-MCA when measuring the blank sample
B_B - counting rate observed in beta-MCA when measuring the blank sample
ε_α – alpha counting efficiency
ε_β – beta counting efficiency.

Since part of the alpha events are classified as beta and vice versa, the observed count rates (A_{0S} and B_{0S}) in each of the two MCA will depend on both alpha and beta decays, according to the relations:

$$A_{0S} = A_T - A_T \cdot X_\alpha + B_T \cdot X_\beta \quad (1)$$

$$B_{0S} = B_T - B_T \cdot X_\beta + A_T \cdot X_\alpha \quad (2)$$

$$A_T = \frac{(A_{0S} - A_B)\cdot(1 - X_\beta) - (B_{0S} - B_B)\cdot X_\beta}{1 - X_\beta - X_\alpha} \quad (3)$$

$$B_T = \frac{(B_{0S} - B_B)\cdot(1 - X_\alpha) - (A_{0S} - A_B)\cdot X_\alpha}{1 - X_\beta - X_\alpha} \quad (4)$$

The gross alpha (Λ_α) and respectively beta (Λ_β) activities of the unknown sample are calculated with the formulas:

Making the necessary substitutions and the correction due to the background, the equations for the true counting rates corresponding to α (A_T) and β (B_T) decays are as follows:

$$\Lambda_\alpha = \frac{A_T}{\varepsilon_\alpha} \quad (5)$$

$$\Lambda_\beta = \frac{B_T}{\varepsilon_\beta} \quad (6)$$

The total alpha activity is determined by considering a counting efficiency of 100% and the total eta activity is determined from beta spectrum by using counting efficiency determined with a calibration standard containing the radionuclides of interest. In the case of the tests carried out in the present work, the counting efficiency determined with ^{90}Sr/Y standard has been used.

Method for simultaneous determination of gross α-β activity using deconvolution of spectrum

An alternate method for pulse discrimination is the spectrum de-convolution. The method is based on the net difference between shapes of alpha and beta scintillation spectra. The method was developed within the Radiation Protection Laboratory of Institute for Nuclear Research and it is presented in detail in the paper *"Simultaneous Determination of Total Alpha and Beta Activity by Liquid Scintillation Spectrometry"* [6]. In short, the method assumes identification in the liquid scintillation spectrum and elimination of the region with contribution from alpha events. The content of spectrum channels is plotted on a graph, excluding the alpha region and spectral data are fitted with a proper function, such as to include in the fitting interval, the alpha region interval. By using an appropriate fitting function, the beta spectrum is generated in the alpha region. For each channel the beta contribution in alpha region is subtracted from the compound spectrum, thus obtaining the deconvoluted alpha spectrum and the alpha count rate is determined from the alpha spectrum.

The total alpha activity is determined by considering a counting efficiency of 100% and the total beta activity is determined from deconvoluted beta spectrum by using the Efficiency Tracing (ET) method.

Results and discussion

Experimental tests for PDD optimization

Two standards have been prepared using ^{241}Am and ^{90}Sr/Y reference solutions. For alpha standard, 1.0046 g of standard solution with the activity concentration of ^{241}Am equal to 570.18 Bq/g was used, while for the beta standard, was used 1.0162 g of standard solution with the activity concentration of ^{90}Sr/Y equal to 256.53 Bq/g. The standard solutions have been prepared in 20 ml polyethylene vials and mixed with 15 ml of scintillation cocktail (Ultima Gold AB). These standards have been counted by LSC for at least 30 min in order to obtain counting statistics with a satisfactory standard uncertainty at varying discriminator settings, where PDD values ranged between 20 and 180. The *tSIE* parameter value for the two standards was about 515.

Figure 2 represents the typical misclassification plot for ^{241}Am and ^{90}Sr/Y standards for determination the optimum setting that result in minimum misclassification of alpha and beta activity. The obtained data show that 102 is the best setting for the PDD.

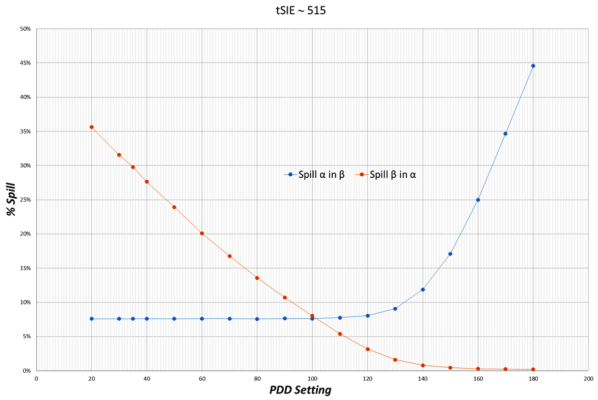


Fig. 2. Optimal PDD value and α/β misclassification factor obtained in ^{241}Am and ^{90}Sr/Y calibration standards - tSIE~515

The experimental tests have been repeated using the same standards for ^{241}Am and ^{90}Sr/Y in which quenching agent was added. The value of the *tSIE* parameter for the second set of measurement was 350. The obtained data shows that the optimal PDD is 78, as can be seen in Fig. 3.

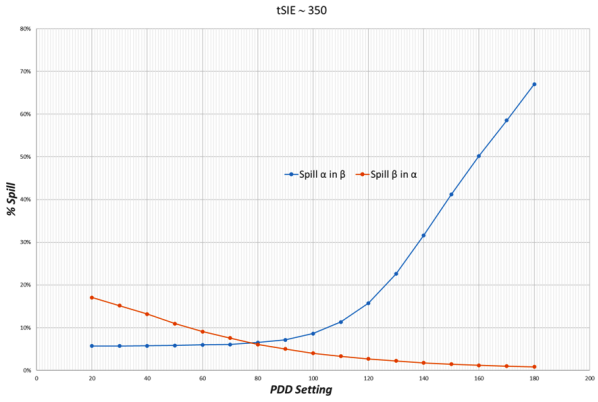


Fig. 3. Optimal PDD value and α/β misclassification factor obtained in ^{241}Am and ^{90}Sr/Y calibration standards - tSIE~350

Experimental tests for the validation of the methods

In order to be able to compare the performances of the two methods, there have been performed experimental tests using two sets of three samples each, containing mixture of alpha and beta emitting radionuclides (^{241}Am and ^{90}Sr/Y) in different proportions. The extinction level of the two sets of samples was different. The activities of the prepared samples and the values of the corresponding *tSIE* parameters are presented in Table 1.

Table 1. Characteristics of samples used for method validation tests

Sample no	tSIE	^{241}Am [Bq/sample]	^{90}Sr [Bq/sample]
Sample 1	515	290.4	132.8
Sample 2		61.0	388.0
Sample 3		509.8	41.2
Sample 4	350	290.4	132.8
Sample 5		61.0	388.0
Sample 6		509.8	41.2

Table 2 summarizes the results obtained by applying the method for simultaneous determination of gross α-β activity using the optimal PDD and the observed discrepancies.

Table 2. Method validation - simultaneous determination of gross α-β activity using the optimal PDD

Sample no	PDD used	^{241}Am [Bq/sample]	^{90}Sr [Bq/sample]	Discrepancies for α measurements	Discrepancies for β measurements
Sample 1	102	312.5	135.2	7.6%	1.8%
Sample 2	102	77.5	377.3	27.1%	-2.8%
Sample 3	102	577.1	37.3	13.2%	-9.3%
Sample 4	78	311.2	131.9	7.2%	-0.7%
Sample 5	78	60.9	400.7	-0.1%	3.3%
Sample 6	78	566.6	37.4	11.1%	-9.3%

Analyzing the obtained results after applying the method of simultaneous determination of gross α-β activity using the optimal PDD, the calculated discrepancies between the theoretical and the measured activity were less than 27.1 % for total alpha and less than 9.3% for total beta.

After the spectrum deconvolution procedure was applied the unfolded alpha and beta spectra were obtained. Figures 4 and 5 there are presented the composite spectra and the unfolded alpha and beta spectra for two of the samples.

Table 3 summarizes the obtain results by applying the method for simultaneous determination of gross α-β activity using deconvolution of spectrum and the observed discrepancies.

Analyzing the obtained results after applying the method of simultaneous determination of gross α-β activity using spectrum deconvolution, the calculated discrepancies between the theoretical and the measured activity were less than 40.6 % for total alpha and less than 8.0% for total beta, respectively.

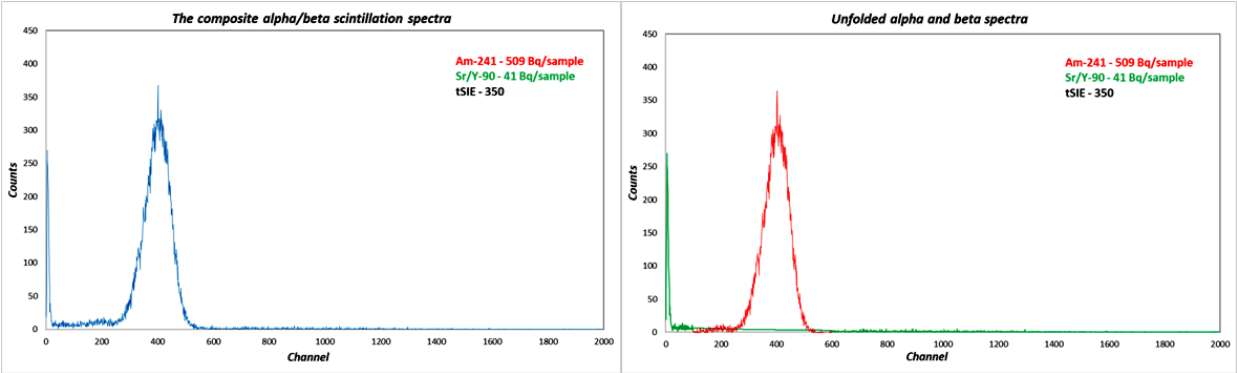


Fig. 4. Alpha-beta spectra (tSIE-350): ^{241}Am ~ 509 Bq/sample, ^{90}Sr/Y ~ 41 Bq/sample

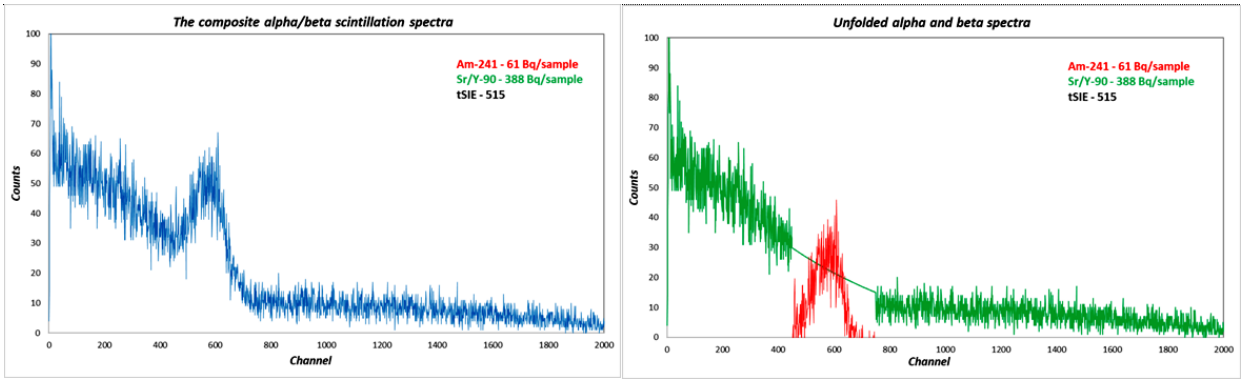


Fig. 5. Alpha-beta spectra (tSIE-515): ^{241}Am ~ 61 Bq/sample, ^{90}Sr/Y ~ 388 Bq/sample

Table 3. Method validation - simultaneous determination of gross α-β activity using the deconvolution of spectrum

Sample no	^{241}Am [Bq/sample]	^{90}Sr [Bq/sample]	Discrepancies for α measurements	Discrepancies for β measurements
Sample 1	277.1	134.2	-4.6%	1.0%
Sample 2	36.2	419.2	-40.6%	8.0%
Sample 3	525.1	38.2	3.0%	-7.3%
Sample 4	279.2	139.3	-3.9%	4.9%
Sample 5	43.8	416.3	-28.1%	7.3%
Sample 6	524.5	38.0	2.9%	-7.7%

Conclusions

The aim of this study was to compare and evaluate the performances of two methods for determining gross alpha-beta activity using LSC.

Although finding the optimum PDD setting could be a difficult job, especially when the radionuclide content of the sample is not known or very complex, analyzing the results obtained after applying the method of simultaneous determination of gross α-β activity using the optimal PDD, the following conclusions were drawn:

- For gross beta measurements, the relative bias is below 9.3%, which give a clear idea of the method's robustness;
- For gross alpha measurements, the relative bias is below 13.2% in most of the cases, except in one sample in which the level of alpha activity is much lower than the beta one.

Analyzing the results obtained after applying the method of simultaneous determination of gross α-β activity using deconvolution of spectrum the highest discrepancies have been obtained in the case of gross alpha determinations for samples in which the level of alpha activity is much lower in relation to beta activity. If the level of alpha activity in the sample is comparable to the beta activity or even higher, relative bias is below 4.6%. For gross beta measurements, the relative bias is below 8.0%, which prove the robustness of the method.

Both methods can be used as screening method in emergency situations, having the benefit of high detection efficiency, simple and fast sample preparation and the possibility of automatic measurement of tens of samples.

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