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Fitting of alpha-efficiency versus quenching parameter by exponential functions in liquid scintillation counting

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ABSTRACT

The objective of this work is to propose an exponential fit for the low alpha-counting efficiency as a function of a sample quenching parameter using a Quantulus liquid scintillation counter. The sample quenching parameter in a Quantulus is the Spectral Quench Parameter of the External Standard (SQP(E)), which is defined as the number of channel under which lies the 99% of Compton spectrum generated by a gamma emitter (^{152}Eu). Although in the literature one usually finds a polynomial fitting of the alpha counting efficiency, it is shown here that an exponential function is a better description.

1. Introduction

In Liquid Scintillation Counting (LSC), the evaluation of factors that contribute to the spectrum is crucial. The sample quenching plays an important role in LSC, such as in alpha and beta counting efficiency, alpha/beta separation and background. Two kinds of quenchings have been identified, the so-called chemical and colour quenching [1]. Although both of them cause essentially the same effect on the measurement, a loss of the number of photons that reach the photomultiplier tube of the detector [2], chemical quenching acts above the production of light from the scintillator, while colour quenching is related to the absorption of light photons once emitted by scintillator.

In the case of Quantulus 1220, the pulse shape analyser (PSA) is used for the separation of alpha-and beta-pulses by measuring their total charge and their delayed fraction of charge released in the photomultiplier, which is different depending on the detected particle (the decay time of the light emissions caused by alpha particles is higher than the decay time for beta particles) [3]. Using standard vials with alpha- or beta-emitters PSA level can be calibrated in order to obtain the best alpha/beta separation, which corresponds to a minimum misclassification. In the case of alpha–beta separation, some authors have reported that both the optimum pulse shape discrimination level and the total interference value between alpha and beta are strongly dependent on the quenching [4–7]. In addition, Alessio et al. [8] found an appreciable increase of the background when the degree of quenching increased. The same has been pointed out by Vaca et al. [9], who have suggested that the background counting rate is affected by the colour quenching in radiation Cerenkov measurements.

On the other hand, Pates et al. [10] have studied the beta-energy dependence on the pulse shape discriminator (PSD) in a Packard Tri-Carb 2550 TR/AB (Packard Instrument Co., Meriden, CT, USA) and they pointed out that a same quench (measured by the tSIE) can differently affect PSD when several quenching agents are compared [11].

Indeed, the observed counting rate depends dramatically on the quenching of the sample, among other aspects. If an unquenched and a quenched samples are compared, the two direct consequences of quenching are lower total count rate in the alpha- and beta-spectra and a shift of counts to the low energy region in the beta-spectrum of quenched sample. As a consequence, a decrease in the counting efficiency in the alpha spectra can be expected due to reduction in the count rate and/or misclassification of alpha pulses in the beta spectra.

Therefore, to correctly measure the activity concentration in a sample and also to improve the alpha/beta separation, a calibration curve of alpha and beta counting efficiency versus an external quenching parameter is required as a routine measurement procedure, especially when environmental samples have to be measured.

In our laboratory we are especially interested in the determination of alpha emitters by LSC after their separation and isolation from different environmental samples (waters, aerosols, biota,

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etc). This implies the selection of a proper PSA value to maximise the alpha counting efficiency and minimise the possible interferences of beta emitters, and to obtain the alpha counting efficiency versus external quench parameter calibration curve for the proper and accurate determination of the alpha emitter under evaluation.

Different mathematical approaches have been reported in the literature for determining the relationship between alpha counting efficiency and each quenching parameter. In most cases the data are fitted to quadratic functions [6,12]. A counting efficiency studied as a function of quenching parameter using a straight-line for very low quenching has also been recently reported [13].

The main objective of this work is to show the goodness of an exponential fitting to evaluate the alpha counting efficiency even in the case of high quenching. The second order polynomial is only a good empirical fitting for low quenching, and additionally it is not theoretically supported. However, in this paper a theoretical base is proposed to support the use of an exponential fitting in the relationship alpha counting efficiency versus quenching parameter.

2. Material and methods

2.1. Materials

20 ml glass vials of low content in potassium (24 ml if the vials are totally full) were used in the experiments. The selected scintillator was OptiPhase HiSafe 3. Samples were traced using 20 μl of a radioactive solution of ^{241}Am. A high purity (99%) carbon tetrachloride (CCl_4) solution was used as quenching agent, considering it as a representative of the quenching agents, which can affect our environmental samples.

The activity measurements were performed using an ultra low level liquid scintillation spectrometer Quantulus 1220. An external capsule of 37 kBq of ^{152}Eu is available at the Quantulus 1220, which was used to measure the Spectral Quench Parameter of the External Standard (SQP(E)), being this parameter the number of channel under which lies the 99% of Compton spectrum generated by the gamma emitter (^{152}Eu) [14]. The Compton dispersion was analysed by using the software OriginPro 8™.

2.2. Experimental procedure

The ^{241}Am traced sample (S_{Am}) was prepared using the following procedure. Firstly 8 ml of distilled H_2O was discharged into the glass vial. Next, 11.18 ± 0.07 dpm of ^{241}Am (20 μl of a standard solution) were added into the water in the vial. The aqueous solution was mixed with 14 ml of scintillation liquid into the vial. Finally, different volumes of CCl_4 were added into the same scintillation solution for each new measurement.

For the sample traced with ^{241}Am, the volume of CCl_4 was successively increased in steps of 20 μl in the first 16 measurements, from 0 to 300 μl of CCl_4. Thereafter the volume of CCl_4 was increased in steps of 50 μl (from 300 to 500 μl), 100 μl (from 500 to 1000 μl of CCl_4) and 200 μl (from 1000 to 2000 μl of CCl_4). The sample was extracted each time from the spectrometer for adding a new amount of CCl_4. In order to assure the homogeneity of the quenched scintillation solution, the vial was always shaken for 3 min after each new addition of CCl_4, and allowed to equilibrate. Otherwise, the quenching agent can remain in a separate phase on the top of the vial, giving wrong values. A total of 30 measurements of the sample traced with ^{241}Am, corresponding to different concentrations of CCl_4, were considered in this experiment. The alpha-counting efficiency for an unquenched sample has been experimentally demonstrated that is nearly constant in the range of high sample volumes (22–24 ml) used for the construction of the alpha counting versus external quenching parameter.

The same procedure was applied to a non-traced sample (S_{Blank}).

The external source of ^{152}Eu, available at the Quantulus 1220 LS, was used for the measurement of the SQP(E) parameter. The SQP(E) parameter corresponding to every volume of CCl_4 was measured 5 times during 1 min each one, in order to assure the reproducibility of the results.

On the other hand, a counting time of 600 min was selected to obtain the counting rate (cpm). The pulse shape analyser (PSA), used to separate the alpha- and beta-pulses, was selected in the threshold level of 105, in order to reduce the alpha-background without the requirement of alpha-interference corrections.

The whole spectra, both the efficiency and the Compton ones for evaluation of the SQP(E), were stored and processed by using the software OriginPro 8™. Fits of the experimental data to mathematical functions, both exponentials and polynomials, were performed using also this software.

The background counting rates were determined using the previously commented non-traced sample, with different concentrations of CCl_4 from 0 to 2000 μl, and in each case were subtracted from the counting rates obtained with the ^{241}Am traced sample in order to determine the net counting rate.

2.3. Determination of normalised efficiency

The normalised counting efficiency, $\varepsilon/\varepsilon_0$, is defined as the ratio between the counting efficiency of a quenched sample and the counting efficiency of an unquenched sample. The normalised counting efficiency is determined in Eq. (1)

$$\frac{\varepsilon}{\varepsilon_0} = \frac{c-b}{c_0-b_0}, \tag{1}$$

where ε is the alpha counting efficiency of quenched samples, ε_0 is the counting efficiency of the unquenched sample, c is the number of counts of traced quenched sample, b is the background for the same amount of quenching agent, c_0 is the number of counts of the traced unquenched sample and b_0 is the corresponding background, using in all cases 600 min for counting time. All these quantities correspond to the alpha spectra.

According to Eq. (1), the measurements of the sample traced with ^{241}Am (S_{Am}) have allowed the determination of the alpha counting efficiency in each case, while the standard of ^{152}Eu is used to evaluate the quenching parameter SQP(E).

The uncertainties of the determined normalised counting efficiencies given by Eq. (1) were calculated by applying Eq. (2), that is

$$\left(\frac{\sigma_{\varepsilon/\varepsilon_0}}{\varepsilon/\varepsilon_0}\right)^2 = \frac{\sigma_c^2+\sigma_b^2}{(c-b)^2} + \frac{\sigma_{c0}^2+\sigma_{b0}^2}{(c_0-b_0)^2} = \frac{c+b}{(c-b)^2} + \frac{c_0+b_0}{(c_0-b_0)^2}, \tag{2}$$

where the uncertainties of the measurements are the square root of the corresponding number of counts ($\sigma_c^2 = c$, $\sigma_{c0}^2 = c_0$, $\sigma_b^2 = b$, $\sigma_{b0}^2 = b_0$).

2.4. Fundamentals for an exponential fit

Because one practical criterion for the quenching evaluation in a sample involves using the value of the SQP(E) parameter, the following step was used to analyse the behaviour of the alpha counting efficiency as a function of SQP(E). The study corresponds to the same samples and measurements in which the counting efficiency was measured.