



Possibilities and limitations of color quench correction methods for gross alpha/beta measurements



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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. Since quenching phenomena are most problematic effect during LSC measurements, the objectives of this paper are to present, consider and evaluate two methods for color quench correction as a proposal for supplement to the ASTM D 7283-06 method for gross α/β determination. The first method is PSA adjustment: application of previously established dependence of optimal PSA discriminator between alpha and beta events on quench level of sample. The second method assumes counting on usual optimal PSA setting and application of previously determined quench calibration curves in order to correct count rates in alpha and beta channels afterwards. Application on obtained activity results of few artesian well water samples and colored spiked samples, based on the measured SQP(E) value of samples, has been demonstrated, as the opportunity to compare the validity, reliability and limitations of both methods. All samples have been counted on low-background liquid scintillation counter Quantulus 1220™.

1. Introduction

Detection by Liquid Scintillation Counting (LSC) offers numerous advantages over conventional techniques: samples do not present self-absorption problems, alpha and beta emitters can be simultaneously measured, counting efficiency for alpha and high energy beta emitters is close to 100%, the determination of low energy beta emitters such as ³H and ²⁴¹Pu is possible, spectral information is provided and sample preparation is usually simple and rapid (in some instances, no radiochemical procedure is required) (Pujol and Sánchez-Cabeza, 1997). Although LSC techniques are ideal for natural radioactivity monitoring of drinking waters, quenching phenomenon appears to be the most problematic effect during LSC detection.

The need for color quench correction had been recognized since natural water samples collected for gross alpha/beta measurements can be significantly quenched. Artesian well waters are not always transparent but yellow and impossible to bleach, which probably origins from the presence of iron and its oxides. Color quench occurrence can cause erroneous results and underestimated activities in screening tests such as gross alpha/beta measurements, therefore quench level of samples is necessary to be determined and its effects on activity determination corrected. Investigations show that color quench affects

more alpha than beta spillover (τ_α increases while τ_β decreases in the presence of increasing amounts of quench agents) (Salonen, 2006).

The main goal of this paper is to present an attempt to correct color quench effects occurring in natural waters analyzed by LSC method. Two methods for color quench correction have been considered and evaluated in this paper, as a proposal for supplement to the ASTM D 7283-06 method [ASTM International D 7283-06] for gross α/β determination. Method 1 is PSA adjustment individually for each sample before counting depending on its quench level, the other method, Method 2, involves additional correction of obtained counts in alpha and beta spectra. Possibilities, advantages and limitations of both methods are discussed.

Measurements were performed by low-background liquid scintillation counter Quantulus 1220™, equipped with Pulse Shape Analysis (PSA) circuit that distinguishes α/β events and stores them simultaneously in different halves of first MultiChannel Analyser (MCA1). The second one, MCA2, generates background spectra into two its halves: coincident and anticoincident with events from radioactive sample. Optimal PSA value has to be experimentally determined as described in (Todorović et al., 2012) and it is known to be dependent on calibration radionuclides' activity concentration, energy and decay schemes, scintillation cocktails and LS counter applied, and quenching agent's

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presence (that can have different quenching strengths, while chemical/color type have different quenching mechanisms) in samples (Stojković et al., 2015; DeVol et al., 2007; Villa et al., 2003). Therefore, the first correction method in case of color quenched samples discussed here, Method 1, is PSA adjustment method – chosen because of the fact that optimal PSA parameter that precisely discriminates alphas from betas presents different value for quenched and unquenched samples. The PSA adjustment according to quench level of samples should provide more accurate generation of alpha and beta spectra.

Quantulus 1220™ measures SQP(E) parameter (Spectral Quench Parameter of the External standard) which is an indication of quench level of a sample. It represents channel corresponding to the 99th percentile of the energy distribution (DeVol et al., 2007) and it is being shifted towards lower channels with quench increment.

Since optimization of alpha/beta spectra separation parameter (PSA) is known to be strongly dependent on quench level, the first method was to readjust PSA setting to corresponding quench indicating parameter (SQP(E)) for each of the counted quenched samples. Alternatively, Method 2 assumed that all samples were counted on usual optimal PSA setting, count rates have been corrected afterwards according to previously determined quench calibration curve.

2. Experimental

Results of experiments were obtained using Ultra Low Level Liquid Scintillation Spectrometer Wallac 1220 Quantulus manufactured by PerkinElmer (Finland, 2002). The spectra were acquired by WinQ and analyzed by EasyView software. Alpha spectrum was analyzed from channels 500–800, while beta spectrum was analyzed from the whole energy region, channels 1–1024.

Standard radioactive sources (^{241}Am , $^{90}\text{Sr}/^{90}\text{Y}$) produced from Czech Metrology Institute, Inspectorate for Ionizing Radiation were used for calibration of the instrument, $A(^{241}\text{Am}) = 37.57 \text{ Bq ml}^{-1}$ with combined standard uncertainty 0.2%, reference date 1/10/2013, $A(^{90}\text{Sr}/^{90}\text{Y}) = 38.18 \text{ Bq ml}^{-1}$ with combined standard uncertainty 0.5%, reference date 1/10/2013. OptiPhase HiSafe 3 was used as a scintillation cocktail, since it is a general-purpose cocktail that handles a broad range of solutes and was available for the research experiments at that moment in laboratory. For the most effective alpha/beta discrimination, Ultima Gold AB cocktail could be used. However, mentioned cocktails, (OptiPhase and Ultima Gold family) do not influence optimal PSA parameter in significant extent (Stojković et al., 2015).

All LSC samples were prepared in 20 ml high performance glass vials (Perkin Elmer). All quenched standards were prepared with 5 ml of active solution and 15 ml of scintillation cocktail (according to the preparation procedure described in [ASTM International D 7283-06]), adding the increasing amounts of yellow dye quenching agent.

Since different types of quench agents affect PSA differently (even when the external quench parameter is the same) (DeVol et al., 2007), yellow dye was used as color quencher as it would probably affect PSA in the most similar way as iron salts in real samples. Preparation of quenched $^{241}\text{Am}/^{90}\text{Sr}$ standards with iron was not possible although it would be most suitable. Added FeCl_3 to spiked water after evaporation formed Fe_2O_3 which could not solve in 0.1 M HNO_3 (which is used according to method [ASTM International D 7283-06]), and sawdust was deposited on the vial walls, so yellow dye was used as a quenching agent instead.

2.1. ASTM methodology and its disadvantages

Water samples were prepared for gross alpha/beta measurements according to ASTM D 7283-06 Standard Test Method [ASTM International D 7283-06]. Samples were always filtered and preserved at the time of collection by adding enough 2 M nitric acid to the sample to bring it to pH 2 or less. Water samples were evaporated to 5 ml,

transferred to a tarred glass scintillation vial using 0.1 M HNO_3 and slowly evaporated to dryness. The residue mass was used as a quench-indicating parameter. Every sample was added with 5 ml of 0.1 M HNO_3 and 15 ml of scintillation cocktail and mixed thoroughly.

Here we give method's calculations according to the ASTM procedure. All the efficiency curves were established by counting of prepared calibration sets of pure alpha (^{241}Am) and beta (^{90}Sr) aqueous standards with different residue masses mixed with liquid scintillator, using the following equations:

$$\varepsilon_{\alpha\alpha} = \frac{R_{\alpha\alpha} - R_{\alpha\beta}}{c_{\alpha} V_{s\alpha}} \quad (1)$$

$$\varepsilon_{\alpha\beta} = \frac{R_{\alpha\beta} - R_{\beta\beta}}{c_{\alpha} V_{s\alpha}} \quad (2)$$

$$\varepsilon_{\beta\beta} = \frac{R_{\beta\beta} - R_{\beta\alpha}}{c_{\beta} V_{s\beta}} \quad (3)$$

$$\varepsilon_{\beta\alpha} = \frac{R_{\beta\alpha} - R_{\alpha\beta}}{c_{\beta} V_{s\beta}} \quad (4)$$

where $\varepsilon_{\alpha\alpha}$ is alpha particle detection efficiency in the alpha ROI, $\varepsilon_{\alpha\beta}$ is alpha particle detection efficiency in the beta ROI, $\varepsilon_{\beta\beta}$ is beta particle detection efficiency in the beta ROI, $\varepsilon_{\beta\alpha}$ is beta particle detection efficiency in the alpha ROI; $R_{\alpha\alpha}$ is count rate of the alpha standard aliquant in alpha ROI, $R_{\beta\beta}$ is count rate of the beta standard aliquant in beta ROI, $R_{\alpha\beta}$ is count rate of the alpha standard aliquant in beta ROI, $R_{\beta\alpha}$ is count rate of the beta standard aliquant in alpha ROI; $R_{\alpha\beta}$ is count rate of the matrix blank contain residue mass approximately equal to the calibration standard in the alpha ROI, $R_{\beta\alpha}$ count rate of the matrix blank counting residue mass approximately equal to the calibration standard in the beta ROI, c_{α} is activity concentration of the reference alpha standard in Bq ml^{-1} , c_{β} is activity concentration of the reference beta standard in Bq ml^{-1} , $V_{s\alpha}$ is volume of the reference alpha standard added to the vial and $V_{s\beta}$ is volume of the reference beta standard added to the vial. Data provided efficiency vs. residue mass curves in (0–500 mg) dry weight interval, which means that, according to the ASTM method, residue mass of sample dictates all the efficiencies, together with misclassification (spillover) factors, calculated as follows:

$$X_{\alpha} = \frac{\varepsilon_{\alpha\beta}}{\varepsilon_{\alpha\alpha}} \quad , \quad X_{\beta} = \frac{\varepsilon_{\beta\alpha}}{\varepsilon_{\beta\beta}} \quad (5)$$

where X_{α} is alpha-to-beta spillover factor and X_{β} is beta-to-alpha spillover factor. The net count rates of samples were calculated as follows:

$$R_{\alpha} = R_{\alpha\alpha} - R_{\alpha\beta} \quad , \quad R_{\beta} = R_{\beta\beta} - R_{\beta\alpha} \quad (6)$$

where R_{α} is net count rate of sample aliquant in the alpha ROI and R_{β} is net count rate of the sample aliquant in the beta ROI.

The corrected alpha R'_{α} count rates and corrected beta count rates R'_{β} (net alpha and beta count rates corrected for spillover) of samples were determined using the following equations:

$$R'_{\alpha} = \frac{R_{\alpha} - R_{\beta} X_{\beta}}{1 - X_{\alpha} X_{\beta}} \quad , \quad R'_{\beta} = \frac{R_{\beta} - R_{\alpha} X_{\alpha}}{1 - X_{\alpha} X_{\beta}} \quad (7)$$

The sample gross alpha activity AC_{α} and sample gross beta activity AC_{β} were calculated from the following:

$$AC_{\alpha} = \frac{R'_{\alpha}}{\varepsilon_{\alpha\alpha} V} \quad , \quad AC_{\beta} = \frac{R'_{\beta}}{\varepsilon_{\beta\beta} V} \quad (8)$$

where V is sample aliquot volume in liters.

Minimum detectable concentration for alpha activity concentration MDC_{α} and for beta activity concentration MDC_{β} were calculated from the equations:

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