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The effect of sample stability on the determination of radioactivity for various radionuclides by liquid scintillation counting



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HIGHLIGHTS

- The variation of quench level over time can be an indication of sample stability.
- The sample stability has impact on α/β discrimination as well as on the counting efficiency.
- Special attention should be paid to the radionuclides with chemical form apt to be adsorbed.
- Shaking a sample and checking the LSC spectra can give the information on physical quench.
- Significant physical quench usually leads to a new peak in the LSC spectrum of an α emitter.

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ABSTRACT

For measuring a sample stored for a long period of time using liquid scintillation counting (LSC), it is necessary to study the long-term stability of the sample. The effect of sample stability on the determination of radioactivity for ^{241}Am , $^{90}\text{Sr}/^{90}\text{Y}$, ^{137}Cs , ^{147}Pm , $^{237}\text{Np}/^{233}\text{Pa}$, and ^3H by LSC has been investigated. The variation of quench level over time can be an indication of sample stability. If the variation in a sample is little, the effect of sample stability on the determination of the above radionuclides can be neglected. Otherwise, the sample stability will have impact not only on the counting efficiency (especially for low energy β emitters), but also on the results of α/β discrimination. For studying the stability of a sample, special attention should be paid to the radionuclides with chemical form apt to be adsorbed, because the quench level of a sample cannot be reflected by the quench index SQP(E) alone when significant physical quench exists. Shaking a sample stored for a long period of time and checking the LSC spectra can give the information on physical quench in the sample. In the range of this study, OptiPhase Hisafe 3 has much better quench resistance than Ultima Gold AB.

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1. Introduction

Liquid scintillation counting (LSC) has been widely used in the field of radionuclide metrology (Feng and He, 2009; Horrocks, 1974; Itoh et al., 2002; Kossert and GrauCarles, 2010; Sanchez-Cabeza and Pujol, 1998), because of its numerous advantages over conventional techniques (Griffin and Sumithrarachchi, 2001; Horrocks, 1974; McKlveen and McDowell, 1984; Sumithrarachchi et al., 2003; Yang et al., 1991, 1994): high counting efficiency, which is close to 100% for α -particles, high-energy ($E_{max} > 200$ keV) β particles or mono-energy (> 50 keV) electrons at low quench levels; simple and rapid preparation of samples; and capability to simultaneously determine α and β emitters; etc.

As reported by Verrezen et al. (2008), there are some cases where LSC vials have to be stored for a long period of time after preparation before being counted or recounted. Another common case is concerning the commercial or homemade quenching standard set, which is usually stored for a long period of time. In such cases, the effect of sample stability on the determination of radionuclide of interest becomes important. The reported researches relevant to sample stability exhibit two problems: (1) Feng et al. (2012) have established the usability of expired cocktails, but not the actual constancy of performance characteristics over time, because they have not actually measured long-term changes in the mixture of cocktail and sample; and (2) the quench effect on sample stability had not been mentioned (Verrezen et al., 2008). Quench is the most important issue for LSC (DeVol et al., 2007; Pates et al., 1998; Sosa et al., 2014; Varlam et al., 2015; Zapata-García et al., 2012), which usually leads to reduction in

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counting efficiency as a result of energy loss in the energy transmission process. According to the stages of the energy transmission, quench can be classified as physical quench, chemical quench and color quench (DeVol et al., 2007). Physical quench occurs with more possibility for samples of long-term studies than for samples of ordinary measurement, because the amount of radioisotopes adsorbed to the wall of the vial increases with time before equilibrium. Therefore, in this study, a series of LSC samples with different quench levels containing various radionuclides (including α emitter, high energy β emitter, and low energy β emitter) were prepared to assess the sample stability which is indicated by the variation of quench level over time.

2. Experimental

2.1. Equipment

An ultra-low background liquid scintillation analyzer (Quantulus 1220) from Perkin-Elmer has been used for the measurements. It is provided with a pulse shape analysis (PSA) function which discriminates α from β radiations and directs them separately into α - or β - multi-channel analyzers (MCA). The discrimination results vary with the PSA parameter, which can be set between 1 and 256. Quenching can be monitored with the aid of the external standard's Compton recoil electron spectrum end point (SQP(E)). The SQP(E) is defined mathematically as the channel number of the external standard spectrum, above which 1% of total intensity of the spectrum is found.

Alpha spectrometer (Alpha Analyst 7200) with passivated implanted planar silicon (PIPS) detectors of 300 mm² active area was from Canberra Industries, Inc.

2.2. Cocktails and other materials

There are many cocktails and vials commercially available, and the quenching behavior of a sample is expected to vary with the kinds of quenching agent (DeVol et al., 2007; Pates et al., 1998; Varlam et al., 2015). In this work, only one or two kinds of frequently used cocktail, vial, and quenching agent were taken as examples to prove that attention should be paid to sample stability if an LSC sample has been stored for a long period of time before measurement.

Two kinds of cocktails were Ultima Gold AB and OptiPhase Hisafe 3 (Perkin-Elmer). Two types of vials were 20 mL polyethylene vials and 20 mL low-potassium glass vials (Perkin-Elmer).

The quenching agent was the simulated solution (referred as SimS) which represents the main composition of Chinese high level liquid waste and reflects the quenching effect of acid and salt. SimS was the mixture of 1.0 mol L⁻¹ HNO₃ and metallic ions, the concentrations of which were 18.3, 6.0, 5.7, 2.9 and 1.5 g L⁻¹ for Na, Fe, Al, Ni and Nd.

Radionuclides ²⁴¹Am, ⁹⁰Sr/⁹⁰Y, ¹³⁷Cs, ¹⁴⁷Pm, and ²³⁷Np/²³³Pa were provided by China Institute of Atomic Energy. It is verified by α spectrometry and LSC spectrometry that the effect of daughter can be neglected for ²⁴¹Am and ¹⁴⁷Pm. The reference solution medium is 0.5 mol L⁻¹ HNO₃ for ²⁴¹Am, 0.01 mol L⁻¹ HNO₃ for ⁹⁰Sr/⁹⁰Y, 0.001 mol L⁻¹ HNO₃ for ¹³⁷Cs, 0.1 mol L⁻¹ HNO₃ for ¹⁴⁷Pm, and 1 mol L⁻¹ HNO₃ for ²³⁷Np/²³³Pa. ³H standard solution (tritiated water, HTO) with the nominal specific radioactivity of 6.234 × 10⁴ Bq g⁻¹ (half-life corrected) was from National Institute of Metrology, China. The nuclear properties of the above radionuclides can be found on Internet: http://www.nucleide.org/DDEP_WG/Nuclides/xx-xxx_tables.pdf, where xx stands for the symbol of the element, xxx for atomic number, e.g. Am-241 for ²⁴¹Am.

Table 1
Preparation of LSC samples containing ²⁴¹Am, ⁹⁰Sr/⁹⁰Y, ¹³⁷Cs or ¹⁴⁷Pm.

Sample ID ^a	Radionuclide(s)	Sample load (μL)	Cocktail	Count rate at the first measurement (cpm) ^b
AB-Am	²⁴¹ Am	10	10 mL Ultima	82,499
AB-Sr/L	⁹⁰ Sr/ ⁹⁰ Y	10	Gold AB	76,188
AB-Sr/H	⁹⁰ Sr/ ⁹⁰ Y	100		761,534
AB-Cs/L	¹³⁷ Cs	20		84,956
AB-Cs/H	¹³⁷ Cs	200		828,308
AB-Pm	¹⁴⁷ Pm	20		96,721
HS-Am	²⁴¹ Am	10	10 mL Opti-	86,474
HS-Sr/L	⁹⁰ Sr/ ⁹⁰ Y	10	Phase Hisafe	76,624
HS-Sr/H	⁹⁰ Sr/ ⁹⁰ Y	100	3	752,503
HS-Cs/L	¹³⁷ Cs	20		85,282
HS-Cs/H	¹³⁷ Cs	200		819,958
HS-Pm	¹⁴⁷ Pm	20		95,807

^a For sample ID, AB stands for Ultima Gold AB, HS for OptiPhase Hisafe 3.

^b Listed are the sum of count rates in both α -MCA and β -MCA (ROI: channels 1–1024, PSA=10). The value of PSA is not important in this case.

2.3. Method

The experiments include three parts.

In the first part, 12 samples containing ²⁴¹Am, ⁹⁰Sr/⁹⁰Y, ¹³⁷Cs or ¹⁴⁷Pm were prepared with two kinds of cocktails in 20 mL polyethylene vials (Table 1). Even though α/β discrimination is not needed for pure α or β emitters, all the 12 samples were measured with α/β discrimination at a series of PSAs (from 10 to 250 with a step of 10) for 5 min, with the aim to study the effect of sample stability on α/β discrimination. The counts in both α -MCA and β -MCA were summed to calculate the count rate for pure α or β emitters. The measurements were repeated as the storage time of LSC samples went on.

In the first part, the optimum PSA parameters were determined with “inflection point” method (Feng et al., 2014a), which features setting the optimum PSA by measuring the sample itself of mixed α/β emitters. However, it can also be applied for a pair of samples of pure α or β emitter, such as the case of this study. In this case, the “inflection point” method includes the following steps: (1) at a series of PSAs, summarizing the count rates of any pair of α and β emitters (with almost the same quench level) within the PK area as Eq. (1), which gives the value of C_{α} ; (2) graphing the relation between C_{α} and PSA, which will always have an inflection point related to the optimum PSA; (3) fitting the data near the inflection point with the function $y = ax^3 + bx^2 + cx + d$, from which the value of optimum PSA can be simply obtained as $-b/(3a)$ when the second derivative of the function, y'' , is equal to zero; (4) calculating the C_{α} at the optimum PSA with the function $y = ax^3 + bx^2 + cx + d$

$$C_{\alpha} = C_{A\alpha}^{PK} + C_{B\alpha}^{PK} \quad (1)$$

In Eq. (1), “C” stands for count rate; the subscript “A” stands for α emitter, “B” β emitter, and “ α ” α -MCA; the superscript “PK” stands for the peak area of α emitter. For example, $C_{A\alpha}$ denotes the count rate of α emitter on the α -MCA. The region of interest (ROI) for any “C” with the superscript “PK” refers to the peak area of α emitter in α -MCA only. The count rate in α -MCA outside the PK area is still assigned to β emitter, because this portion of count rate will not interfere with the count rate of α emitter.

In the second part, no new sample was prepared. We just re-measured the seven samples A1 to A7, which had been measured using the same LSC analyzer in our previous study (Feng et al., 2014b). The samples A1 to A7 were prepared with Ultima Gold AB in 20 mL polyethylene vials, containing 0.0328, 0.0629, 0.1055, 0.2095, 0.3129, 0.4190, and 0.5256 g solution of ²³⁷Np/²³³Pa,

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