Systematic study of particle quenching in organic scintillators

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ABSTRACT

Among the different factors that affect measurements by organic scintillators, the majority of attention has been focused on those related to the scintillator (i.e., ionization, chemical, color and optical quenching), and less attention has been paid to the loss of energy before the particle (i.e., alpha or beta) arrives at the scintillator (i.e., particle quenching). This study evaluates the effect of particle quenching in different scintillation methods (i.e., using two plastic scintillation microspheres (PSm1 and PSm2), liquid scintillator and gel scintillator) by measuring solutions that contain increasing concentrations of NaCl, $BaCl_2$ and glycerin. The results show the importance of particle quenching in PSm measurements because detection efficiency decreases with increasing concentrations of the quenching component, although the spectrum position and external standard parameter remain constant. The results have shown evidence of particle quenching, although at a lower magnitude, in the liquid scintillation or gel scintillation measurements. Moreover, the use of two PSm with different diameters and salty compound that alters the equilibrium of the liquid and gel emulsions also exemplified the importance of the transmission of optical photons through different scintillation media (i.e., optical quenching). Improvement and deterioration of the optical conditions on the scintillation media is manifested as a movement of the spectrum to higher and lower energies, respectively. The results obtained with PSm were confirmed by Monte Carlo simulation.

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1. Introduction

The development of organic scintillators began at the end of the 40 decade with the aim to use these compounds to transform the kinetic energy of electrons that are emitted by radionuclides into optical photons that could be detected by photomultipliers [1–3]. During the following years the developments were focused in: the development of organic scintillators in different physical states (e.g., crystal, liquid, gel, or plastic), the formulation of new scintillators, the establishment of the main properties of scintillators [4] and the mechanism(s) involved in these properties [5], and the development of analytical applications that can be used to determine radioactivity [6]. Liquid scintillation, LS, was early established as the best technique for determining and quantifying beta and alpha emitters. During the last twenty years, main developments have been guided by the necessity to find selective, efficient and reproducible analytical procedures, and a few studies have focused on a better understanding of some of the mechanisms that are involved in the scintillation process [7–9].

The detection efficiency of organic scintillators depends on the efficiency in which the kinetic energy of the particle (i.e., alpha or beta) is transformed into optical photons and is detected by photomultipliers. This transformation can be divided into three consecutive stages:

A. The path of the particle from the point in the solution where the disintegration is produced, through the medium, and until the particle interacts with the scintillator. In this stage, energy can be lost due to excitations or ionizations of the molecules of the medium (e.g., water or sample compounds).

B. Transformation of the energy that is deposited in the organic solvent molecule into photons that are emitted by the secondary scintillation solute. In this stage, energy can be lost by the excessive density of the solvent molecules excited (i.e., ionization quenching [10]), the excitation of non-scintillating molecules (i.e., chemical quenching [11]) or other inefficient processes (e.g., autoabsorption).

C. Travel of optical photons from the scintillator, through the medium and the detection chamber, until they reach the PMT surface. In this stage, energy and photons can be lost due to absorption by colored samples (i.e., color quenching [12]) or by inefficient photon transmission processes [13].

Although a large number of studies have focused on understanding and calibrating the effects of the two last stages, little information exists on the effects on the measurement by the

processes that cause energy losses before the particle arrives at the scintillator. In this sense, the use of Plastic Scintillation microspheres, PSm, provides a new opportunity to understand the phenomena that cause the transformation, partially or not, of the kinetic energy of a particle into detectable scintillation photons. PSm are a solid mixture of organic fluorescent molecules in a completely polymerized solvent (e.g., polystyrene or polivinyltoluene) and have a size that is in the tens or hundreds of microns. The aqueous sample can be mixed with the PSm to obtain a heterogeneous scintillation medium, which presents some advantages over LS (e.g., Scintillation proximity assay [14], continuous measurements [15,16], non-production of mixed waste [17–19] and others [20,21]).

It can be assumed that the scintillation mechanism of PSm must be similar to that of LS, but previous experiments [22] have demonstrated that the loss of energy during the path of a particle through the sample until it arrives to the scintillator is of high importance because it causes an important decrease in the detection efficiency for weak beta emitters (e.g., ^3H). However, for high-energy beta emitters, detection efficiency values are similar to those of LS. Moreover, it has also been observed that efficiency decreases with an increase in the diameter of microspheres [23].

These experimental findings, which were observed when measuring with PSm, can be compared with Monte Carlo simulation of the transport of electrons through the aqueous phase before they reach the scintillator. During the last decades and in parallel to the increase in computing power, different codes have been successfully developed to simulate the track of the alpha and beta particles and gamma rays in different media. One example is the Penelope [24,25] software, which was developed to simulate electrons and gamma radiation in a wide range of energies (from a few eV to MeV) and in geometries of different sizes (from micrometers to meters). This software has been successfully applied in the measuring of radionuclides with PSm in a Triple to Double Coincidence Ratio detector (TDCR) [26].

The aim of this work is to evaluate and quantify the effect of different solutions (e.g., salty and dense solutions) on measurements with PSm and, by comparing the results with those of other organic scintillators, i.e., liquid scintillators and gel scintillators, to determine a general mechanism for particle quenching in organic scintillators. To fulfill this objective, we have determined the experimental detection efficiencies when measuring ^3H and ^{36}Cl with PSm of different diameter, liquid scintillator and gel scintillator. Values obtained with both PSm were compared to those obtained by the Monte Carlo simulation using a modification of the Penelope software. In a second stage, we have evaluated the effect of variation in the detection efficiency with the variation of the concentrations of different compounds (e.g., NaCl, BaCl$_2$ and glycerin) on measuring ^3H and ^{36}Cl when using different PSm (PSm1 and PSm2), liquid scintillator and gel scintillator.

2. Experimental

2.1. Reagents and solutions

All reagents that were used were analytical or scintillation grade and deionized water was used to prepare the solutions.

Detec (Ottawa, Canada) supplied the plastic scintillation microspheres (Polystyrene UPS-89). The PSm were separated by sieving, and two different PSm, which were named PSm1 (which was segregated with a sieve with pores between 120 μm and 180 μm) and PSm2 (which was segregated with a sieve with pores between 400 μm and 500 μm), were used in the experiments. OptiPhase Supermix (liquid scintillator) and Insta-Gel Plus

(gel scintillator) Scintillation Cocktails were supplied by PerkinElmer (Waltham, MA. USA). Measurement solutions were prepared in 6-ml Pico Prias polyethylene vials (PerkinElmer).

A ^3H, active stock solution of 350.8 ± 5.8 Bq/g was prepared from a standard that contained 3589 ± 58 Bq/g of ^3H (Eckert–Ziegler, Berlin, Germany) in water; and a ^{36}Cl, active stock solution of 2.22 ± 0.03 Bq/g, which was prepared from a standard that contained 22.24 ± 0.26 Bq/g of ^{36}Cl in a water solution that contained 65 μg/l of NaCl (from CERCA/LEA, Pierrelatte, France), were used to prepare the measurement solutions.

NaCl (from Merck, Darmstadt, Germany), BaCl$_2$ and Glycerin (from Panreac, Castellar del Vallés, Spain) were also used to prepare the measurement solutions.

2.2. Apparatus

An AJ2-HS centrifuge (Beckman-Coulter Inc., Brea, USA) was used to centrifuge the measurement vials, and an Ultrasons-P ultrasonic bath (from JP Selecta, Abrera, Spain) was used to apply ultrasonic waves to the solutions.

A 1220 Quantulus liquid scintillation spectrometer (PerkinElmer) was used to measure the count rate and the energy spectrum of the PSm, LS and GS samples. The detector has two photomultipliers to detect the scintillation pulses of the samples and two photomultipliers to detect the scintillation pulses caused by the cosmic rays in the active guard. The signals coming from the sample detector that are not in coincidence with a signal in the active guard detector (true sample signals) are stored in a 1024 channel multichannel analyzer in function of the height of the pulse (energy spectrum). Correlation between channel numbers and pulse height of the pulses (energy of the pulse)) is logarithmic and unknown for the end user. As a result for each sample measurement we obtain the number of pulses detected (coincident counts that are recordered), the real time of measurement and the energy distribution of the pulses detected.

A LS 13 320 single-wavelength laser diffraction particle size analyzer (Beckman-Coulter Inc., Brea, USA) was used to determine the probability size distribution of the plastic scintillation microspheres that were used (i.e., PSm1 and PSm2).

2.3. Sample preparation

Measurement samples were prepared by adding 2 g of the organic scintillator (i.e., PSm1 or PSm2 plastic scintillation microspheres, liquid scintillator or gel scintillator) and 1 ml of the counting solution (i.e., active or blank) to a 6-ml polyethylene vial. After preparation, the vials were shaken using ultrasonic waves for 2 min and then centrifuged for 10 min at 83.3 Hz to ensure homogenization and to improve the efficiency and reproducibility of the experiments.

The counting solutions were prepared by diluting a known quantity of the active stock solution (^3H or ^{36}Cl) and the corresponding quenching agent (NaCl, BaCl$_2$ or Glycerin) into a known volume of the carrier solution to obtain the desired activity and concentration of the quenching agent in the measurement sample.

The activity of the measurement samples was around 350 Bq for the ^3H solutions and 2.17 Bq for the ^{36}Cl solutions.

The measurement samples contained an increasing concentration of the various quenching agents (NaCl, BaCl$_2$ or Glycerin). The concentrations of the measurement samples that contained NaCl were as follows: 0 M, 0.3 M, 0.7 M, 1.4 M, 2.1 M and 2.6 M. The concentrations of the BaCl$_2$ measurement solutions were as follows: 0 M, 0.05 M, 0.10 M, 0.15 M, 0.2 M, 0.25 M, 0.3 M, 0.7 M and 1.4 M. Finally, the glycerin solutions had the following percentage of weight values: 0%, 14.42%, 28.85%, 43.33%, 57.75%, and 72.17%.