

NUMBER OF CATHODE ELECTRONS PER ABSORBED ENERGY IN LIQUID SCINTILLATION COUNTERS, DERIVED FROM RESOLUTION OF RADON SPECTRA

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ABSTRACT. An important parameter in a liquid scintillation (LS) event is the ratio of the number of electrons released at the cathode of a photomultiplier tube divided by the energy lost by the electron causing the fluorescence scintillation. We term this ratio the “scintillation figure of merit” (SFM), and it depends both on the scintillation cocktail and the system used. High values enhance the counting efficiency of low-energy beta emitters and the energy resolution of alpha spectra. When the SFM is known, the counting efficiency of a dissolved beta emitter with a known energy spectrum can be calculated. It is difficult to measure this parameter using the modern 2-tube coincidence systems, but easy with the flexible, single-phototube system described. A simple method is outlined to determine the SFM with useful accuracy for any LS system based on the energy resolution of the pulse-height spectrum of ²²²Rn and its decay products. A system independent function is also presented, describing the relationship between the SFM of a toluene scintillator and a readily determined Rn resolution parameter. The Rn samples are easy to prepare for all scintillators.

INTRODUCTION

An important parameter in a liquid scintillation (LS) event is the ratio of the number of electrons (N_{ce}) released at the cathode of a photomultiplier tube (PMT) divided by the energy E (keV) lost by the electron causing the fluorescence scintillation. This ratio is constant above ~ 100 keV, but decreases somewhat at lower energies because of ionization quenching (Horrocks 1964, 1979; Theodórsson et al. 2003).

A high ratio enhances both the counting efficiency of low-energy beta emitters like ³H and ¹⁴C, and the energy resolution of alpha particle spectra. This parameter has therefore frequently been called “figure of merit” (e.g. Gibson and Gale 1968), a term that is ambiguous as it is widely used to describe the sensitivity of low-level counting systems. We have therefore chosen to call the electron/energy ratio the scintillation figure of merit (SFM):

$$SFM = N_{ce}/E \quad (1)$$

Knowing the SFM allows one to calculate the counting efficiency of any dissolved beta emitter with a known energy spectrum (e.g. Grau Malonda 1999; Theodórsson 2006) and to compare the merits of LS systems.

For a given system, SFM depends on: 1) the photon yield of the scintillator; 2) the vial/PMT light collection efficiency; and 3) the cathode photon sensitivity, which is similar for all PMTs used in modern LS counters. SFM is proportional to each of these factors (Theodórsson 2006). Horrocks (1964, 1979) pioneered in the measurement of SFM. It is difficult to measure it directly using coincidence systems, but relatively easy using our flexible, laboratory-made, single-PMT detector unit.

In spite of the importance of the SFM in LS metrology, its value is never given in specifications for commercial LS systems. Their SFM, however, can be derived from the absolute counting efficiency of tritium (e.g. Coursey et al. 1986), which is always given in the specifications, but unfortunately without specifying the type of scintillator used.

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We demonstrate how SFM can be derived with useful accuracy for any LS system with a selected scintillator from an easily determined parameter that describes the energy resolution of the pulse-height spectrum of the ^{222}Rn series. The parameter, defined below, depends on the level of overlapping of the closely spaced alpha peaks of ^{222}Rn (5.49 MeV) and ^{218}Po (6.00 MeV).

EXPERIMENTAL

Figure 1 is a schematic diagram of the simple experimental LS detector unit used, which is inside an aluminium tube. A 3-mL dome-shaped sample vial sits on top of a 28-mm-diameter PMT, Hamamatsu 6094, clamped vertically. The vial is wrapped (except for the bottom facing the PMT) with 2 layers of thin Teflon[®] tape, which give high light reflection. This reflective arrangement and closed-vial/phototube geometry gives high scintillation photon collection at the PMT cathode (Theodórsson 2005). In our system, this efficiency can be enhanced by a thin layer of glycerol between the bottom of the vial and the PMT, resulting in optical coupling.

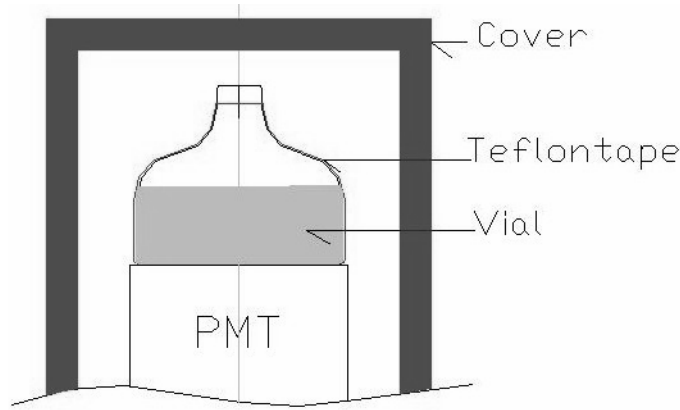


Figure 1 The LS detector unit

A stabilized high-voltage supply operates the PMT at 400–800 V. The anode pulses are fed into a low-noise preamplifier and then into a linear amplifier where the amplification can be varied by a factor of 2 in 6 steps, from 4 to 128. The amplified pulses are sent to a 512-channel multichannel analyzer (MCA). This number of channels matches the resolution of the Rn spectra well.

RESULTS

Determination of SFM

We mainly use 3 mL of toluene to which we add 45 mg butyl-PBD. Its SFM value is determined in the following way. The size of a scintillation pulse, $P(E)$, generated when an electron deposits energy E keV in the scintillator, is proportional to the average number of electrons, $N_{ce}(E)$, released at the cathode of the PMT:

$$P(E) = P_{1e}N_{ce}(E) \quad (2)$$

where the proportionality constant, P_{1e} , is the average contribution to the pulse size of each of the $N_{ce}(E)$ cathode electrons. The pulse size $P(E)$ is measured here by its corresponding MCA channel number. From Equations 1 and 2, we get:

$$SFM = ((P(E))/P_{1e}) \quad (3)$$

The value of SFM is determined in 2 steps. First, P_{1e} is determined, followed by the ratio of pulse size and deposited energy, $P(E)/E$, i.e. channels per keV.

P_{1e} is measured in the following way. Single electrons are emitted spontaneously from the photocathode, each releasing a varying number of secondary electrons at the first dynode, typically 5 on average. The spectrum of these small pulses is measured at maximum amplification, *128, and at a PMT anode voltage where the small pulses rise well above the amplifier noise level (728 V). The pulse-height spectrum is characterized by a broad peak (Figure 3), which, with good approximation based on theory, can be described by a Poisson distribution. The size of a pulse is measured by its channel number in the multichannel analyzer. P_{1e} corresponds to the channel number at the maximum of the cathode single-electron peak, which here occurs at channel number 62.1 (Figure 2).

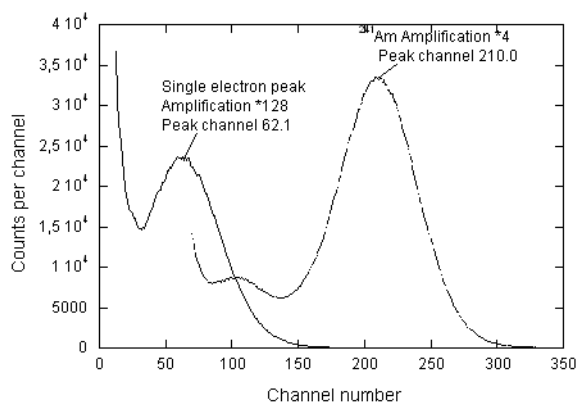


Figure 2 The pulse-height spectrum of single cathode electrons and ^{241}Am .

In the second step, we determine $P(E)/E$. The pulse size, $P(E)$, is proportional to the deposited energy, except at energy well below 100 keV, where it is reduced by ionization quenching (Horrocks 1979; Theodórsson et al. 2003). We use the 59.5-keV gamma peak of an external ^{241}Am source to find the value of $P(E)/E$, measured at the same high voltage as used above, 728 V, but at 32 times reduced amplification (i.e. at *4). The middle of the peak occurs in channel number 210 (Figure 2). Inserting appropriate values into Equation 3, we get: $\text{SFM} = 210 \times 32 / (62.1 \times 59.5) = 1.82$ cathode electrons per keV.

As the measurements of the Rn resolution presented below are made at lower PMT anode voltage (550 V), we measured SFM from 700 (where the cathode single electron pulses rise sufficiently above the amplifier noise level) to 900 V. The results (Figure 3) show that SFM varies only a little with anode voltage in this higher voltage range; thus, we assume that the SFM is practically the same at 550 V as at 728 V.

Relationship Between SFM and Energy Resolution of ^{222}Rn Spectrum

A high SFM value brings us improved alpha spectrum resolution, which is usually measured by the full width at half maximum (FWHM) of a mono-energetic peak (Knoll 1989). For our case, we propose a different easily determined parameter, radon resolution (RnR), defined below, which is derived from the spectrum of ^{222}Rn and its 4 short-lived decay products (Table 1). The Rn sample is made by bubbling through the scintillator 5 mL of air from a syringe taken from the space above 10 mL of ^{226}Ra water solution in a 20-mL vial.

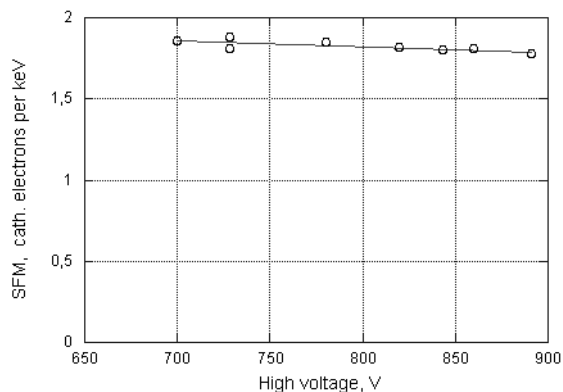


Figure 3 SFM as a function of high voltage

Table 1 The short-lived ^{222}Rn series.

Nuclide	Half-life	Particle, energy (MeV)
^{222}Rn	3.82 d	α 5.49
Rn		
^{218}Po	3.05 min	α 6.00
RaA		
^{214}Pb	26.8 min	β 0.65
RaB		
^{214}Bi	19.7 min	β 3.17 (23%), 1.75 (77%)
RaC		
^{214}Po	0.16 ms	α 7.69
RaC'		
^{210}Pb	22 yr	β 0.018
RaD		

The Rn spectrum (Figure 4) has 3 alpha peaks, due to ^{222}Rn , ^{218}Po , and ^{214}Po (Table 1), superimposed on the continuous beta spectra of ^{214}Pb and ^{214}Bi . The peaks of ^{222}Rn and ^{218}Po are generally strongly overlapping. As a measure of the Rn energy resolution, we define RnR as the ratio of the count rate, T , at the top of the ^{222}Rn peak to the count rate, V , in the valley between the alpha peaks of ^{222}Rn and ^{218}Po (Figure 4):

$$RnR = T/V \quad (4)$$

RnR is a sensitive measure of change in SFM.

All the measurements of RnR discussed below were made at 550 V anode voltage, well below the value where SFM was determined (728 V). To further investigate its voltage dependence in a lower voltage range, we determined RnR, which depends on SFM, at varying anode voltage, within the limits that the variation in amplification allowed (Figure 5). It shows that RnR (and therefore also SFM) is nearly constant in this range. We therefore assume that the value of SFM at 550 V is close to that at 728 V.

The aim of this study is to find the relationship between this RnR and SFM. For this reason, the photon collection (and thus the SFM) must be varied. We do this by inserting a varying number of thin

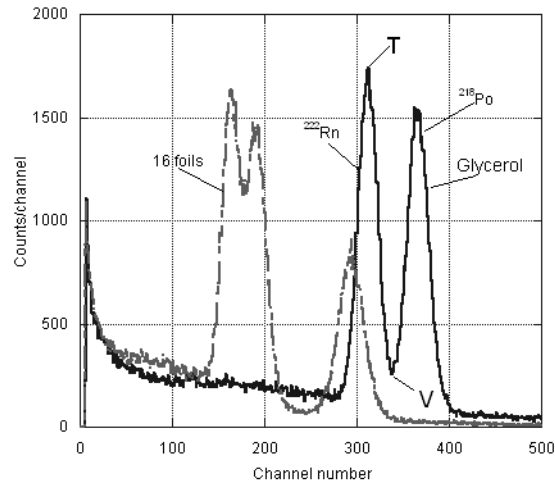


Figure 4 Radon pulse-height spectra for toluene scintillator with glycerol and with 16 polyethylene foils respectively between vial and PMT.

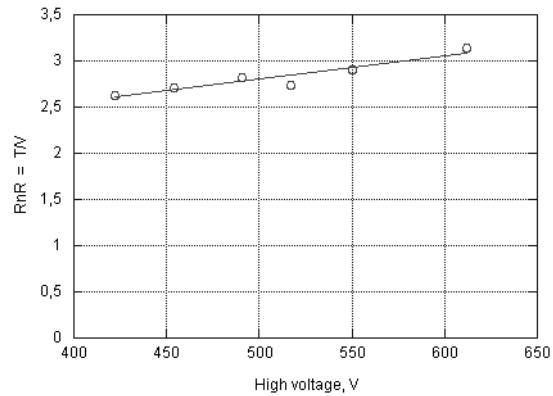


Figure 5 RnR as a function of high voltage

polyethylene foils between the bottom of the vial and the face of the PMT. Each new foil reduces the pulse height by ~6%. To find the SFM also at higher energy resolution, we also measured the Rn spectrum with a thin glycerol layer between the vial and the PMT, as this reduces reflection losses. Figure 4 shows the Rn pulse-height spectra at 2 different levels of energy resolution: 1) with glycerol and 2) with 16 foils between the bottom of the vial and the PMT.

At each of these settings, SFM can be derived from its measured value with no foil and the shift in pulse size (measured by the channel of the mid-²²²Rn peak) as SFM is, at fixed high voltage and amplification, proportional to the pulse size according to Equation 3. Figure 6 shows the calculated SFM value as a function of measured RnR. Once the RnR has been measured for a LSC system with a toluene scintillator, the system's SFM can be derived from this graph.

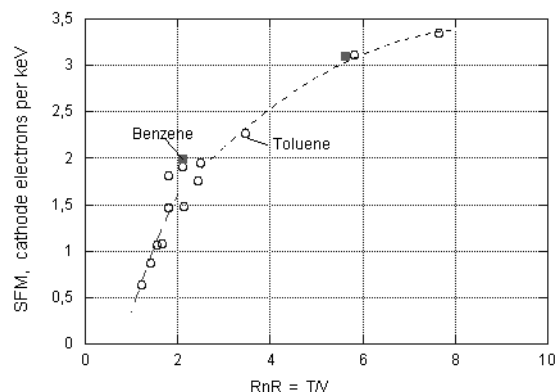


Figure 6 SFM as a function of RnR

The Rn spectrum with glycerol between the vial and the PMT demonstrates the high resolution of the system. It is therefore suitable for the measurement of alpha emitters.

CONCLUSIONS

We hope that this study will lead to further work, e.g. comparing measured ^3H and ^{14}C counting efficiency with theoretically calculated values based on the SFM value derived from RnR, and a study of the relationship between RnR and SFM for other scintillators, vials, and geometries.

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