

# DETERMINING FUNDAMENTAL PARAMETERS OF A SINGLE-PHOTOTUBE LIQUID SCINTILLATION COUNTER

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**ABSTRACT.** After 50 yr of liquid scintillation (LS) counting, basic principles still play a minor role in studies and applications of the technique and the empirical approach usually dominates. We hope the present study will be a step towards changing this attitude. Using a laboratory-made, flexible, single-phototube system, the value of 2 basic parameters has been determined from the pulse-height spectra of 1) single-photocathode electrons; 2) Cerenkov radiation; and 3) fluorescence; using the calculated cathode photon efficiency. The 2 parameters include a) the photon collection efficiency of the vial/phototube unit, which depends only on the counting system used; and b) the photon scintillation yield, which depends only on the scintillation cocktail. From these 2 parameters, other parameters are derived. Furthermore, the present study demonstrates the advantage of single-tube detector units.

## INTRODUCTION

The foundations of the liquid scintillation (LS) counting technique were established late in the 1950s. In the early systems, the faint light flashes were detected by a single photomultiplier tube (PMT). Due to noise pulses from its cathode, these systems had a high background pulse rate in the counting windows of tritium and radiocarbon, the 2 most important radiotracers. This problem was solved in the mid-1950s with the introduction of the 2-tube coincidence system, where a detected signal was only registered when pulses appeared simultaneously in both tubes.

During the early period of the technique, the study of fundamental LS parameters was technically difficult. In the late 1970s, PMTs had improved significantly, and the LS systems had better and more sophisticated electronics that usually included a multichannel analyzer (MCA). Modern 2-PMT coincidence systems with fixed amplification and high voltage are, however, not suitable for fundamental LS studies. This situation can explain the empirical nature of various LS studies where basic LS parameters are not taken sufficiently into account; the following are just a few examples. The tritium counting window is sometimes given as 0–18 keV, but the lowest energy beta particles detected under the best circumstances have a lower threshold at 3–4 keV, corresponding to a maximum counting efficiency of ~65%. The tritium counting efficiency is sometimes used to compare different scintillation cocktails. It is, however, much simpler to compare the Compton edge of the pulse-height spectra of the system's internal gamma source, which gives an equivalent comparison. In Cerenkov counting, the pulse size is sometimes given in keV, which is meaningless. The logarithmic amplification of many systems is a barrier to basic understanding as the energy corresponding to each channel is not known, making comparison of results from different laboratories difficult.

I have used a laboratory-made, flexible, single-PMT system to quantitatively measure basic LS parameters. This study demonstrates at the same time the advantage of the single-PMT system and a linear energy scale compared to a logarithmic scale.

## BASIC STEPS IN SCINTILLATION DETECTION

In the first step of the scintillation process, an electron with energy  $E$  produces  $N_{sp}$  photons (fluorescence or Cerenkov). Of these photons,  $N_{cp}$  hit the cathode of the PMT:

$$N_{cp} = QLN_{sp} \quad (1)$$

where  $Q$  is the quench factor and  $L$  is the photon collection efficiency. In the following, I assume that there is no quenching, i.e.  $Q = 1.0$ . The photon collection efficiency is an important LS parameter that depends only on the vial/PMT geometry.

A fixed fraction of these  $N_{cp}$  photons release an electron at the cathode. The number of the cathode electrons,  $N_{ce}$ , depends on the sensitivity (quantum efficiency),  $\varepsilon$ , of the cathode:

$$N_{ce} = \varepsilon N_{cp} \quad (2)$$

These electrons are the beginning of the electric signal. After internal amplification in the PMT and in the LS system's amplifier, the electrons give an output pulse,  $P$ , that is proportional to  $N_{ce}$ :

$$P = P_{1e} N_{ce} \quad (3)$$

The proportionality factor  $P_{1e}$  is the average contribution of each of these  $N_{ce}$  electrons to the pulse  $P$ , which is measured by its MCA channel number. Combining Equations 1–3, we get:

$$P = N_{sp} L \varepsilon P_{1e} \quad (4)$$

This equation will be discussed in detail below.

## COUNTING MODES

For measuring beta active radioisotopes, LS systems are used in 2 different modes: 1) fluorescence counting, where the radioactive sample is dissolved in a scintillation cocktail; and 2) Cerenkov counting, where the sample is dissolved in water. In the latter mode, which can only be used for high-energy beta emitters ( $>0.5$  MeV), the counting efficiency is much lower, but there is less or no interference from low-energy emitters, sample preparation is generally simpler, and the radioactivity can after measurement be recovered for other studies.

Samples often contain only 1 radioisotope with a known beta energy spectrum. In each counting mode, it is then enough to know the value of only 1 system parameter to be able to calculate, with the help of theory, the counting efficiency: 1) the photon collection efficiency for Cerenkov counting, and 2) the number of cathode electrons emitted per keV of absorbed electron energy for fluorescence.

## APPARATUS

Figure 1 is a schematic diagram of the simple, flexible LS detector unit used in this study. It has a single 28-mm-diameter, flat-end-face PMT (Hamamatsu 6094) clamped vertically. A dome-shaped, 3-mL quartz counting vial sits on top of the tube. The vial is wrapped (except for the bottom, facing the PMT) with 2 layers of a thin Teflon<sup>®</sup> tape, which gives high light reflection. The vial/PMT unit is inside an aluminium tube.

A stabilized high voltage supply operates the PMT at 500–800 volts. The anode pulses are fed to a low-noise preamplifier and then to a main linear amplifier, where the amplification can be varied in 6 steps by a factor of 2, from 4 to 128. The pulses are then sent to a 512-channel MCA. This number of channels has been selected as it matches the spectral resolution of the system well.

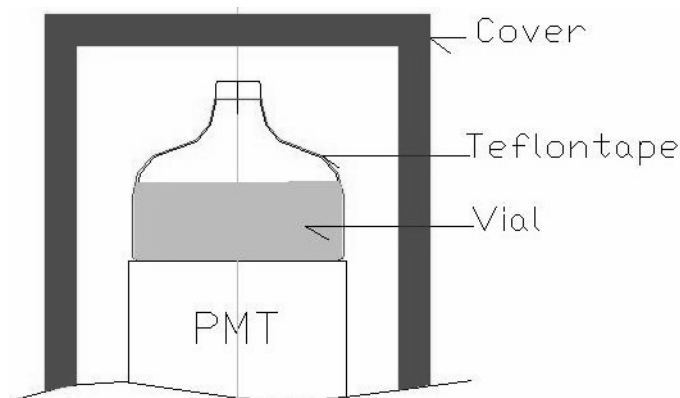


Figure 1 Schematic diagram the LS detector unit

### BASIC LS PARAMETERS

The performance of an LS detector with a selected sample can be described by 3 fundamental parameters, as mentioned above:

1. Average PMT cathode photon efficiency,  $\epsilon$ ;
2. Photon collection efficiency,  $L$ ; and
3. Fluorescence scintillation efficiency,  $Sf$ , for organic liquid scintillators.

When these parameters are known, others can be found.

#### The Average PMT Cathode Photon Efficiency, $\epsilon$

The quantum efficiency is a parameter that describes the response of the PMT cathode to photons hitting it.  $P(\lambda)$  is the probability that a photon with wavelength  $\lambda$  will release an electron from the cathode, which depends on its wavelength. The probability (or sensitivity) curve is provided by the producer of the PMT (Figure 2). We are, however, only interested in the mean (or wavelength-weighted) efficiency of all the photons,  $\epsilon$ . The mean efficiency depends both on the type of cathode material and the photon spectral distribution, which can be fluorescence or Cerenkov radiation.

The average number of electrons,  $N_{ce}$ , released from the cathode in a scintillation event is then:

$$N_{ce} = \int I(\lambda)N_{cp}P(\lambda)d\lambda \tag{5}$$

where  $N_{cp}$  is the number of photons in the sensitive wavelength window of the PMT falling on its cathode in a scintillation event, and  $I(\lambda)$  describes their spectral distribution (Figure 2). The integration is taken over the wavelength interval for which the cathode is sensitive, from 300 to 650 nm (Figure 2). This gives the average, or wavelength-weighted, photon efficiency of the cathode:

$$\epsilon = N_{ce}/N_{cp} \tag{6}$$

The scintillation event can either be fluorescence in a scintillation cocktail or Cerenkov radiation in water, each with its characteristic spectral distribution,  $I(\lambda)$  (shown in Figure 2).

As the quantum efficiency curve and the relative intensity of the photons as a function of wavelength are known,  $\epsilon$  can be calculated. Using the curves shown in Figure 2, we find through numerical inte-

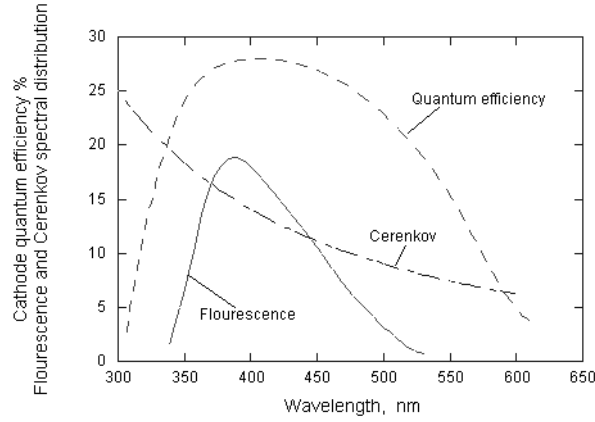


Figure 2 PMT spectral quantum efficiency and spectral distribution of Cerenkov and toluene fluorescence radiation.

gration that for toluene scintillator,  $\varepsilon_{fl} = 0.22$  electrons/photon, and for Cerenkov radiation in water,  $\varepsilon_{Ce} = 0.15$  electrons/photon. It can be assumed that all modern PMTs used in LS detectors give practically the same efficiency.

#### The Photon Collection Efficiency, $L$

The photon collection efficiency  $L$  is the fraction of the total number of photons ( $N_{sp}$ , fluorescence, or Cerenkov photons) produced in a scintillation event that are collected on the cathode of the PMT:

$$L = N_{cp}/N_{sp} \quad (7)$$

It is reasonable to assume that  $L$  is practically the same for fluorescent and Cerenkov scintillation events for the same detector unit, although there may be a small difference due to a difference in refraction index of water and scintillator, which affects photon loss.  $L$  is a measure of how well the designer of the system has succeeded in minimizing the loss of photons in the vial/PMT unit.

The value of  $L$  is determined here through a study of the peak produced by cathode single electrons and through Cerenkov spectra. The size of a pulse,  $P$ , is proportional to the number of electrons,  $N_{ce}$ , released at the cathode of the PMT in a scintillation event:

$$P = P_{1e}N_{ce} \quad (8)$$

where the proportionality factor  $P_{1e}$  is the average contribution of each of the  $N_{ce}$  cathode electrons to the pulse size. By combining Equations 6–8 we get:

$$L = (P/P_{1e})/\varepsilon N_{sp} \quad (9)$$

The key to the determination of  $L$  is that it is possible to theoretically calculate the number of Cerenkov scintillation photons produced in water by an electron of known energy (Knoll 1989), which in this study are either maximal energy beta particles of a known radionuclide, or Compton edge electrons produced by an external gamma source.

The value of  $L$  is determined in 2 steps. First,  $P_{1e}$  is determined. 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 63.7 (Figure 3). At minimum amplification, \*4, the peak would be 32 times lower, corresponding to channel number 1.99. It is interesting to note that at above 3 times this level, at channel number 190, there is practically no interference from cathode single-electron pulses.

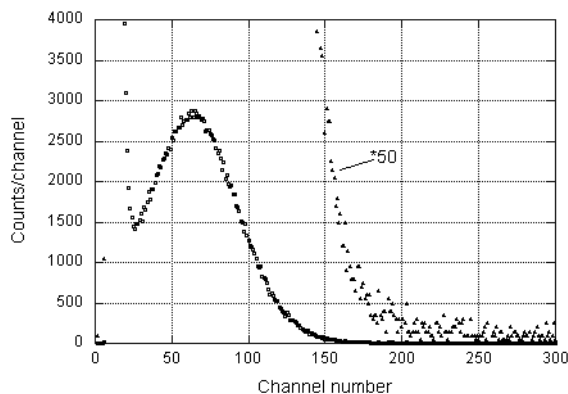


Figure 3 Pulse-height spectrum of cathode single electrons measured at 728 V and amplification \*128.

Six Cerenkov spectra were subsequently measured at minimum amplification (\*4), but at the same anode voltage (728 V)—3 produced by beta-particles and 3 by Compton electrons released in the water by external gamma radiation (Table 1).

Table 1 Cerenkov measurements, high voltage 728 V, amplification \*4.

Nuclide	Max energy (keV)	Cerenkov photons	Cathode electrons
<sup>90</sup> Sr β	546	41	5
<sup>90</sup> Y β	2273	748	66
<sup>40</sup> K β	1320	320	31
<sup>22</sup> Na γ 1275 keV	1062	208	28
<sup>60</sup> Co γ 1333 keV	1119	232	26
<sup>208</sup> Tl γ 2614 keV	2381	781	70

Figure 4 shows one of the Cerenkov spectra, that of Compton electrons of <sup>208</sup>Tl and other radionuclides in the <sup>232</sup>Th series, produced by an external thorium oxide gamma source. It should be noted that the x axis scale is here the number of released cathode electrons (1.99 cathode electrons per channel), as this is the most natural way to display Cerenkov spectra.

From the Cerenkov spectra, we read the pulse size corresponding to the most energetic electrons through extrapolation to the background count rate based on plausible guesswork. The accuracy can no doubt be significantly improved by supporting the extrapolation by theory.

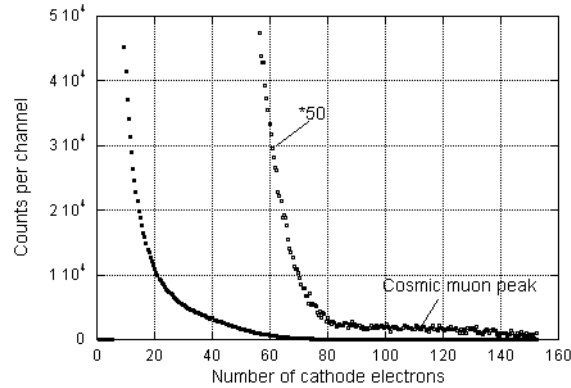


Figure 4 Cerenkov pulse-height spectrum of the  $^{232}\text{Th}$  series

The number of Cerenkov photons produced by an electron of energy  $E$  can be calculated theoretically (Knoll 1989). The number of photons,  $N_{sp}$ , in the wavelength interval from 300 to 600 nm as a function of electron energy is shown in Figure 5.

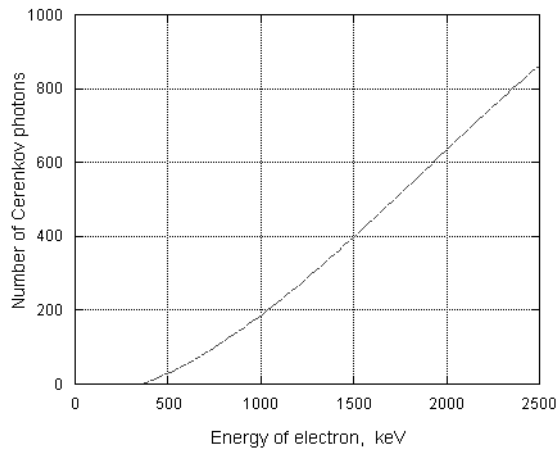


Figure 5 Number of Cerenkov photons produced by electrons in water as a function energy.

Results of these measurements are given in Table 1. Figure 6 shows number of cathode electrons as a function of the number of Cerenkov photons. The slope of the line is 0.091 cathode electrons per scintillation photon. According to Equation 5, the slope is equal to  $\epsilon L$ , which gives a photon collection efficiency of  $L = 0.61$ . The detector unit collects 61% of the scintillation photons on the cathode of the PMT.

A thin layer of glycerol between the bottom of the vial and the face of the PMT provides an optical contact between these 2 surfaces and increases the photon collection. Cerenkov measurement with air and glycerol between the vial bottom and PMT, respectively, showed that glycerol increased the photon collection by 32% compared to air, or to  $L = 0.80$ . This is apparently the first direct determination of this important parameter. In the literature, estimated values ranging from 10 to 100% can be found.

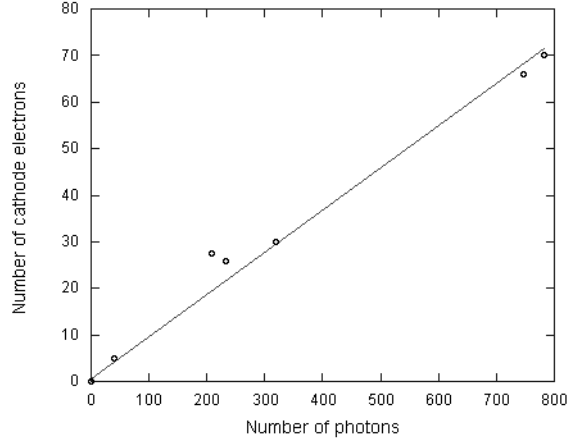


Figure 6 Number of cathode electrons as a function of the number Cerenkov photons.

The Cerenkov counting efficiency for a high-energy beta emitter, dissolved in water, can now be calculated. A beta particle of energy  $E$  produces in average  $N_{sp}(E)$  photons (Figure 5) and these release  $N_{ce}$  cathode electrons, where, according to Equations 1 and 2,  $N_{ce}(E) = L \times \varepsilon \times N_{sp}(E)$ . The average probability (DP) of detecting this pulse is

$$DP = 1 - e^{-N_{ce}(E)} \quad (10)$$

The Cerenkov detection efficiency ( $\varepsilon_{Cer}$ ) is then (Mann et al. 1991):

$$\varepsilon_{Cer} = \int P(E)(1 - e^{-N_{ce}})dE \quad (11)$$

where  $P(E)$  is the normalized Fermi distribution function. The lower integration limit corresponds to the beta energy that gives a pulse that rises sufficiently above the amplifier noise level and the upper limit is the maximal energy of the beta spectrum.

### Fluorescence Scintillation Yield, $S_p$

The fluorescence scintillation yield,  $S_p$ , is the number of fluorescence photons,  $N_{sp}$ , emitted in a scintillation event per unit energy (keV) deposited by an electron in the scintillation cocktail used:

$$S_p = N_{sp}/E \quad (12)$$

Equations 6–8 then give:

$$S_p = N_{sp}/(L\varepsilon_{fl}E) = (P/E)/P_{1e}/(L\varepsilon_{fl}) \quad (13)$$

where  $S_p$  is a characteristic parameter for the scintillation cocktail used. We use air-saturated toluene to which we add 15 g butyl-PBD per liter.

To find the value of  $P/E$ , the fluorescence spectrum of an external  $^{241}\text{Am}$  gamma source (59.5 keV) was measured at the same high voltage as used above (728V) and at an amplification of \*4. The middle of the 59.5-keV peak was in channel number 210 (Figure 7). The top of the single cathode elec-

tron peak was 62.1 at amplification \*128, i.e. at 32 times higher amplification. Inserting appropriate values into Equation 6, we get:  $S_p = (210/59.5) / (62.1/32) / (0.61/0.22) = 14.5$  photons/keV.

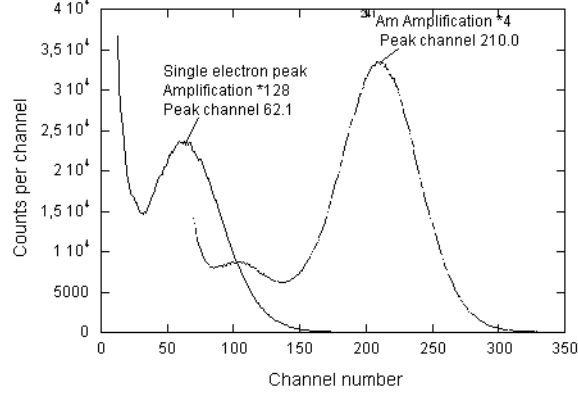


Figure 7 Peaks of cathode single electrons (amplification \*128) and  $^{241}\text{Am}$  (amplification \*4), both measured at 728 V.

### Other LS Parameters

Using the value of the 2 basic LS parameters that are now known, the value of other parameters can be determined. The scintillation figure of merit (SFM) is the ratio of the number of cathode electrons,  $N_{ce}$ , and the energy,  $E$  (keV), deposited in the scintillator by an electron (Mann et al. 1991):

$$SFM = N_{ce}/E = (P/P_{1e})/E \quad (14)$$

This equation shows that neither the value of  $L$  nor  $\varepsilon$  need to be known to find the SFM. Inserting measured values, we get for the air saturated toluene scintillation cocktail:  $SFM = 210 / [(62.1/32) \times 59.5] = 1.82$  cathode electrons per keV.

This is the LS parameter that has highest practical value. Knowing the SFM, it is possible to calculate the detection efficiency of a beta emitter dissolved in the scintillator. The average number of cathode electrons resulting from the emission of a beta particle of energy  $E$  (keV) is  $E \times SFM$ . The detection efficiency,  $\varepsilon$ , is then:

$$\varepsilon = \int P(E)(1 - e^{-L \cdot SFM \times E})dE \quad (15)$$

where  $P(E)$  is the normalized Fermi distribution function and the integration is from the lower to the upper energy limits of the selected energy window. The lower limit of integration is usually determined by the wish to reduce background and interference from a radionuclide of lower energy.

The energy scintillation efficiency,  $S_E$ , is the fraction of deposited energy,  $E$ , that is converted to light in a fluorescence scintillation event:

$$S_E = N_p E_{ph}/E = S_p E_{pa} \quad (16)$$

where  $E_{pa}$  is the average energy of the emitted photons,  $E_{ph}$ , which is close to 3.2 eV. Inserting the values found above, we get:  $S_E = 14.5 \times 3.2 \cdot 10^{-3} = 0.046$  photon energy/total energy; 4.6% of the electron energy deposited in the toluene cocktail is thus converted into light energy. This value is in

acceptable agreement with that found by Skarstad et al. (1968), who got a 4.2% for de-aerated benzene with p-terphenyl solution, which probably has scintillation efficiency similar to the air-saturated toluene used here. Skarstad et al. used a complex method, which is, however, independent of the sensitivity curve of the PMT.

## CONCLUSIONS

The values of 2 basic parameters of the liquid scintillation (LS) counting technique: 1) the photon collection efficiency of the vial/PMT unit, and 2) the fluorescence photon scintillation yield (average number of photons per keV deposited by an electron in the scintillator) have been determined by direct measurements. From these values, 2 further parameters are derived: the scintillation figure of merit and the energy scintillation efficiency.

The present study should be considered as a step toward a better understanding of LS parameters and their application in practical work. It may lead to further work that will bring theory closer to experimental data, for example, comparing measured  $^3\text{H}$  and  $^{14}\text{C}$  counting efficiency with theoretically calculated values, based on SFM, and comparing theoretically calculated Cerenkov counting efficiency with measured values for selected radioisotopes.

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