The measurement of scintillation emission spectra by a coincident photon counting technique

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Abstract. In the evaluation of novel scintillators, it is important to ensure that the spectrum of the light emitted by the scintillator is well matched to the response of the photomultiplier. In attempting to measure this spectrum using radioactive sources, it is found that so few photons are emitted per scintillation event that conventional spectroscopic techniques cannot easily be used. A simple photon counting technique is presented, using two photomultipliers operated in coincidence, the one viewing the scintillator directly, while the other views it through a monochromator. This system allows the spectrum to be measured without using specially cooled photomultipliers, intense radioactive sources or particle beams.

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1. Introduction

One of the most important parameters of any novel scintillator is the spectral response of the light produced. For the scintillator to be experimentally useful, this response must match the responses of readily available photomultiplier photocathodes or other photodetectors.

In the case of organic scintillators, the ultra-violet induced fluorescence spectrum is frequently measured, as this is considered to be identical to the scintillation spectrum [1, 2, 3] since both emissions are due to the same electronic transitions. However, it remains difficult to study the ultra-violet induced spectrum accurately where the absorption and emission bands of the fluorescing molecules overlap. Further, the spectra emitted by organic solutions can be considerably different from the the spectra obtained by direct UV-excitation of the pure solutes [3].

For inorganic scintillators, the absorption bands are generally of such short wavelengths that generating a suitable excitation spectrum becomes problematic and transmission of this through more than a few millimetres of material impossible [4]. Additionally, in many inorganic crystals, the emission spectra produced are known to depend markedly on the exciting particles [5].

These considerations suggest that it would be preferable to excite the scintillator directly with radiation, rather than to rely on the ultra-violet induced spectrum. In the case of organic scintillators, since α-particles will only penetrate the surface layers and because there are problems with absorption of γ-rays in small samples, previous measurements have mainly been done using β-particles [6, 1, 2]. However, with the β-particle energies emitted by available radioactive sources and typical organic scintillator efficiencies (one photon for 100eV energy deposited [7]), very little light is produced per event. This light must be fed through a monochromator and then passed to a broad band photon detector in order to derive the spectrum.

The scintillation photons are emitted isotropically and the angular acceptance of monochromators is inevitably low, consequently only a tiny fraction of the photons produced will reach the detector. There is also the trade-off that if the monochromator is set for higher resolution, then the throughput is unavoidably reduced and the signal-to-noise ratio is degraded. Attempts have been made to avoid the problems encountered with β-sources by using electrons from accelerators [8, 9]. While these were successful, the equipment required is unlikely to be generally available. More recent work to characterize a range of inorganic compounds has made use of a bench-top pulsed X-ray system [10].

The measurement of scintillation emission spectra is essentially a signal-to-noise ratio problem. The signal (here, the number of photons per event) can be increased by depositing more energy in the scintillator. Clearly there is a limit to the energy obtainable from β-particles from radioactive sources, while the use of accelerators to produce higher energy electrons unnecessarily complicates the measurement. Increasing the rate of events and integrating over many events is a workable option but this involves

working with excessively radioactive sources and, while this may have been an option in the past, is no longer acceptable.

Since it is clearly difficult to increase the signal, the next obvious approach is to attempt to reduce the noise. The principle source of noise is that of single-electron noise in the recording photomultiplier. Cooling the photomultiplier reduces this, but since not all the noise is of thermal origin, this approach can only be partially successful. The present method uses two photomultipliers and a coincidence technique to suppress the noise, whatever its physical origin.

2. Previous Measurements

Before describing the coincidence method in detail, it is worth reviewing the previous measurements and comparing their capabilities and achievements.

Ott et al. [6] used a photographic technique to integrate the faint scintillation light from organic liquid samples over a long period of time. A 3.7GBq ^{137}Cs source providing β-particles of 0.51 and 1.17MeV was used. The resolution appears to have been less than 5nm although this is not specified directly. Clear inconsistencies between the fluorescence and scintillation spectra were observed.

Van Sciver [5] examined the spectral emission of sodium iodide crystals, excited by either a ^{60}Co γ-ray source or a ^{242}Cm α-particle source. The light was passed through a monochromator onto a photomultiplier cooled with dry ice and the DC output of this was fed via an amplifier to a pen recorder. No specification of the spectral resolution of the system is given but responses are presented which suggest that the resolution is 5nm.

Birks [1] compared the scintillation and fluorescence spectra of organic solutions. A grating spectrometer was coupled to a photomultiplier, the DC output of which was coupled to a chart recorder to chart the spectrum. β-particles of 1.71MeV were provided by a 52MBq ^{32}P source. To overcome the low intensity of the light emission, the input and output ports of the spectrometer were widened, giving a resolution of 8nm for the better scintillators and 16nm for the poorer materials.

Langenscheidt [2] further studied organic solutions, providing a comparison of spectra obtained by UV excitation with those obtained from α, β and $\gamma-$radiation. The output of the spectrometer was coupled to a photomultiplier cooled to $-80°$C, the DC output of which was displayed on a chart recorder. Three radioactive sources were used, a 37MBq ^{241}Am source providing 5MeV α-particles, a 19MBq ^{247}Pm source providing 225keV β-particles and a 74GBq ^{192}Ir source providing 0.2–1.36MeV γ-rays. In the case of the β source, the particle energy was so low that the range in the liquid was only ~ 0.1mm. The resolution appears to have been around 5nm although, again, this is not directly specified.

Horrocks [11, 8] used intense pulses of 3MeV electrons derived from a Van de Graaff generator. These were typically 3ns long and with beam currents of 400mA. The test samples were organic liquid samples, flame sealed in quartz ampoules so that

they could be held in a vacuum line for exposure to the beam. The light produced was passed through a monochromator and onto a photomultiplier, individual pulses being recorded. A resolution of 0.7nm was attained. Problems were encountered both with short term quenching and with long term damage of the scintillator as a result of the intense radiation flux.

Pronko et al. [9] developed a system in which intense pulses of 20keV electrons, produced by a simple electron gun, were used to excite plastic scintillator samples held in a vacuum line. The light was fed through a spectrometer and onto a photomultiplier. The signal-to-noise ratio was improved by use of a lock-in amplifier which modulated the electron beam. The resolution was 3nm but problems were encountered because the electron beam current was so high that deterioration of the light output of materials was observed.

Moses et al. [10] designed a facility for the bulk evaluation of novel inorganic scintillators using a pulsed X-ray tube. Each 100ps long pulse produced, on average, 40 X-ray quanta per steradian with a mean energy of 18.5keV. The X-rays excited crystal or powdered inorganic samples and the light from these was monitored by a photomultiplier. By placing a monochromator between the sample and the photomultiplier, the emission spectrum of the scintillator could be determined. A resolution of 12nm was achieved. The system was limited by the low level of light.

Kirov et al. [12, 13] examined the spectral emission of water-equivalent plastic and liquid scintillator solutions. A 1.7GBq ^{90}Sr/^{90}Y source was used, providing β-particles of 2.28MeV. The light from the scintillator was fed through a monochromator and onto a photomultiplier. The output from this was integrated over a few tens of seconds and an identical time interval dark background signal was subtracted. The monochromator accuracy was measured as 0.8nm though the step size used in actual measurements was 2.5nm. Both the monochromator and the data acquisition were computer controlled allowing the automatic measurement of spectra.

3. Coincident Photon Counting

In the coincident photon counting technique, two photomultipliers are used to increase the signal-to-noise ratio of the measurement. The arrangement is shown schematically in figure 1. One of the photomultipliers, denoted the "total-event" tube, views the scintillator sample directly, while the other, denoted the "monochromated" tube records only photons that have passed through the monochromator. The signal from the total-event photomultiplier is passed to a discriminator set to trigger at a typical β-particle flash, while the output from the monochromated photomultiplier is passed to a discriminator set to detect single photons. The coincidence of the two discriminator outputs is taken and the rate of coincidences is measured in real time. In this way, photons recorded by the monochromated tube are only counted if they are in coincidence with a scintillation flash recorded by the total-event photomultiplier. This effectively suppresses the single-photon noise of the monochromated photomultiplier.
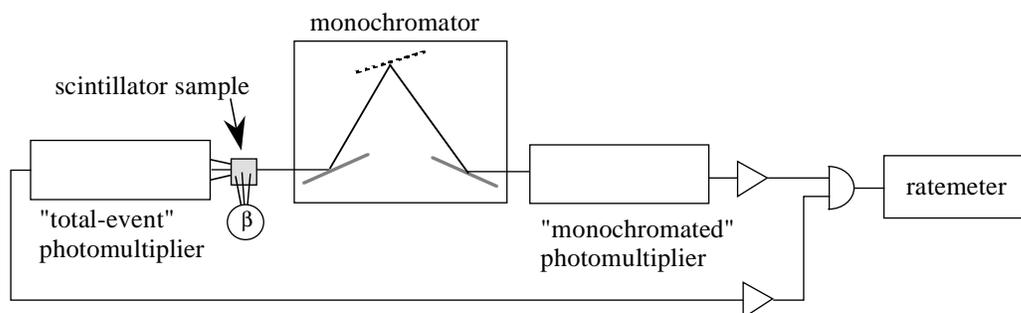
Figure 1. The experimental setup

The system was designed to study both liquid and plastic scintillator samples. Liquid samples were contained in $10 \times 10 \times 55$mm disposable acrylic cuvettes while plastic samples were machined to similar dimensions. The samples were arranged so that β-particles entered the scintillator from below through the base. The β-particles had an end-point energy of 2.28MeV and were obtained from a 3.7MBq ^{90}Sr source. This source was held in a brass collimator consisting of a 50mm long 20mm diameter cylinder with a central cavity. A 1mm diameter axial hole was drilled into this cavity in order to provide a well-defined β-particle beam with negligible exposure in other directions. The sample was arranged so that the volume of scintillator immediately above the base was aligned with the input slit of the monochromator. The base of the acrylic cuvettes was
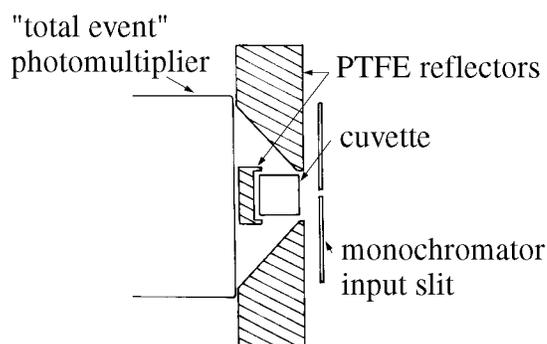
Figure 2. Plan view of the arrangement of the sample and PTFE reflectors

1mm thick, so some absorption of β-particles in the wall could be expected. However, at 2.27MeV, the range in material of density 1.0 is about 10mm so the attenuation effect is negligible. Tests with cuvettes containing only water or pure mineral oil indicated there was no problem with scintillation in the walls of the cuvette. Before liquids were measured they were sparged with solvent-saturated nitrogen to remove dissolved oxygen.

The monochromator was an Applied Photophysics High Radiance Monochromator, selected purely on the grounds of availability. Coupled to the output port of the monochromator was an Electron Tubes 9829QA quartz windowed photomultiplier, chosen to ensure that the spectral response of the measuring system extended into

the ultraviolet.

A system of diffuse reflectors fabricated from PTFE (polytetrafluoroethylene) ensured that, while as much light as possible entered the input port of the monochromator, some of the light was directed to a second photomultiplier, the "total-event" photomultiplier, here an Electron Tubes 9954KA. This arrangement can be seen in plan view in figure 2. For clarity, the radioactive source and its collimator are not shown; they are immediately below the sample cuvette.

The anode signals of the photomultipliers were both fed to discriminators. The "total-event" tube had an Ortec 584 with the discriminator set to about 100 photo-electrons while the "monochromated" tube had a Mechtronics 511 photon discriminator set at about 0.3 times the single photon level. The outputs of the discriminators were set to about 50ns width and fed to a LeCroy 466 coincidence unit. The coincidence output was simply counted over 100 second periods with a Racal 9902 counter timer and this rate gave a measure of the intensity of the scintillation light at the wavelength selected by the monochromator.

With the resolution of the monochromator set at 5nm, and typical organic scintillators observed at peak wavelength, the rate of coincidences was a few tens per second. This meant that statistically significant results could be obtained with count periods of 100s per 5nm wavelength bin. A complete spectrum from 360nm to 540nm would take a little over an hour to complete. The system described here was manually controlled but it would be simple to automate the measurement by motor driving the monochromator. It was also possible to improve the resolution at the expense of longer counting times.

In order to determine the relative spectral response of the recording system, tests were performed using light from a quartz-halogen lamp and a mercury vapour lamp. The light from these was attenuated with a series of pinholes and fed through the monochromator and onto the quartz window of the photomultiplier. For these tests, the single-photon count rate was determined directly without a coincidence requirement. By varying the monochromator over the range of interest and comparing the single-photon rate with the known spectral emission of the sources, a set of correction curves for the instrumental response of the monochromator and quartz window photomultiplier were obtained. In the case of the mercury vapour lamp, individual emission lines could be discerned, and this provided a check on the wavelength calibration of the monochromator. One series of the correction measurements was performed with a sample wall from one of the cuvettes inserted between the light source and the monochromator to ensure that the corrections included wall losses.

With the photomultiplier used and with the organic scintillators being studied, the correction factors were minor. If the technique were used for materials emitting further into the ultraviolet or into the green, where the response of the photomultiplier is reduced, the accuracy with which the corrections were known would begin to dominate the measurements.

Since it was clear at an early stage that the coincident photon counting technique

provided excellent signal-to-noise ratio, no effort was made to cool the photomultipliers. Indeed, it is not obvious that any improvement to the measurements would be made by such cooling.

4. Results

To show typical results from the system, spectra from two liquid scintillator samples, XLS169A and XLS169C produced by Zinsser Analytic (UK) Ltd., are presented in figure 3. Both materials are ternary scintillators using POPOP and bis-MSB. XLS169A
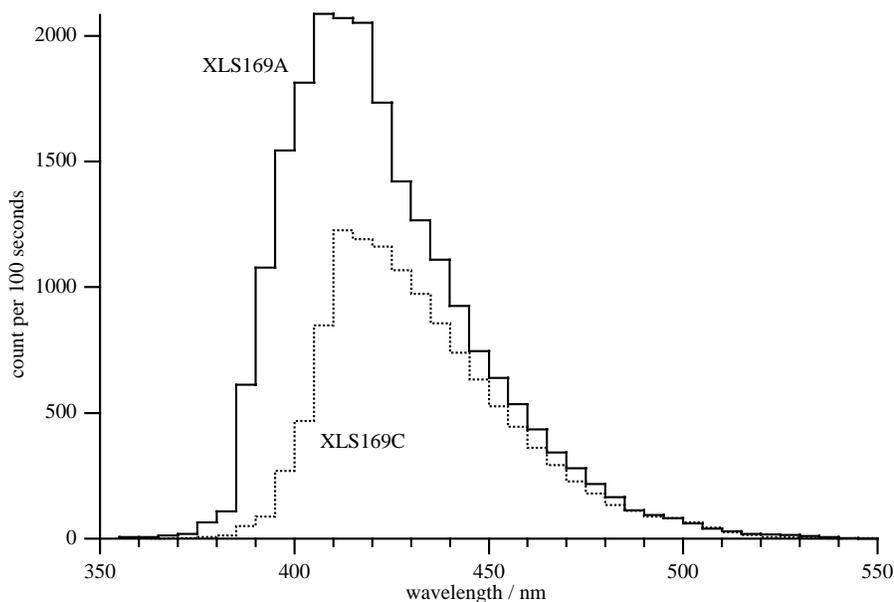
Figure 3. Typical liquid scintillator spectra

is based on diisopropylnapthalene, whereas XLS169C is a mineral oil and naphthalene mixture. It can be seen that while the two spectra are similar on the long wavelength side, the XLS169C cuts off more sharply on the short wavelength side, presumably because the mineral oil solvent has lower transparency in the near ultraviolet.

It is worth noting that the integral of each curve gives the total amount of light emitted over all wavelengths and hence provides a comparative measure of efficiency for the scintillator sample. In the work of Kirov et al. [12], the corrections needed for accurate calculation of the number of photons per unit wavelength are considered and a detailed error analysis for estimating the scintillator efficiency from spectral measurements is given.

5. Conclusions

The coincident photon counting technique is demonstrated as a simple method of determining scintillator spectral response. It requires only low cost equipment typically available in laboratories where scintillators are studied. The apparatus described uses

a relatively low activity β-particle source and organic scintillator samples, but the technique could easily be extended to use α or γ sources and modified for inorganic or even cryogenic scintillators.

In the case of ternary organic scintillators, employing a solute and a wavelength shifter, care must be taken that the light paths through scintillator are longer than the extinction length of the wavelength shifter. If this is not the case, unshifted photons will escape the volume and the measured spectrum will be inaccurate. For a typical commercial plastic scintillator, Bicron BC400/NE-102, this has been measured [14] as 120μm. With the geometry described above and materials with realistic levels of wavelength shifter, no problems were encountered.

In the case of inorganic scintillators with long decay times and more than one decay component, it might be possible to examine changes in spectral output as a function of time by using a short coincidence gate and specifying a time delay from the total-event discriminator.

The system could further be extended to examine the emission spectra of wavelength shifter materials by using a radioactive source to excite a scintillator which is arranged to illuminate one of the large faces of a rectangular slab of polished acrylic. The wavelength shifter can either be doped into the acrylic or be deposited as a film on the large face. If light from the slab is coupled through one of the small edges into the monochromator, it will be uncontaminated by the original scintillation light spectrum.

The coincident photon counting technique described may also be applied to the measurement of the spectra of any other pulsed light emission such as Cherenkov light, plasma or spark discharges.

Acknowledgments

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