Chapter 5

Two-Parameter Pulse Height Analysis in Liquid Scintillation

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INTRODUCTION

Virtually all liquid scintillation spectrometers since 1950 have utilised dual-phototube coincidence detectors to reduce background count rate.[1] Various methods of combining the output signals from each detector have been used for pulse height discrimination against background.[2–7]

This chapter will show how analysis of the relative pulse height from each detector is utilised to provide additional information about the sample specimen and photodetectors. Two-parameter pulse height analysis[8] is used to examine the relative response from each photodetector. Correlations between the response from each detector are used to resolve some of the ambiguities which arise in single-parameter observations. The added dimension provides a new means of discriminating between background events and those caused by scintillations in the sample, by rejecting regions of the 2-parameter plane. Various transformations of the 2-parameter spectrum into a single-parameter spectrum to improve background discrimination, spectral resolution and/or efficiency will be described.

OPERATIONAL OVERVIEW OF A COINCIDENCE SPECTROMETER

A generalised coincidence spectrometer is depicted in the block diagram of Fig. 1. Pulses from each of the photodetectors are simultaneously applied to a high-speed (typically less than 50 ns) coincidence detector, and also to a processing element H which transforms the 2-parameter X and Y pulse height information from each phototube into a single-parameter pulse height h. A pulse height analyser (PHA) sorts and counts the pulses. The coincidence detector enables pulse height analysis only when a signal is received from both detectors at the same time ($\sim$50 ns). Gating by the coincidence detector can alternatively be accomplished prior to the H transform, i.e. analogue gating.

The H processing element can be analogue or digital. For example, H may be a voltage, current or pulse width proportional to the magnitude of h, or it may be a serial or parallel digital word proportional to the magnitude of h. More than one transform can be used. Depending upon H, the pulse height analyser can utilise

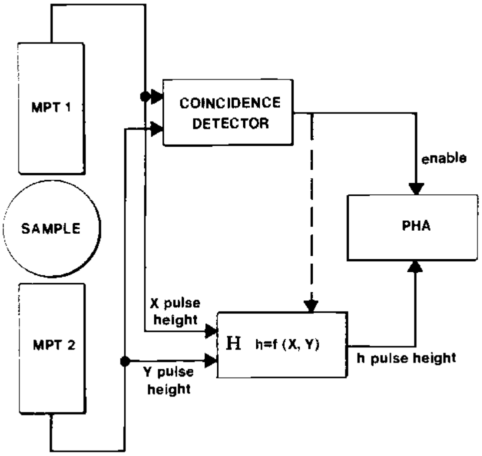

Fig. 1. Generalised coincidence spectrometer.

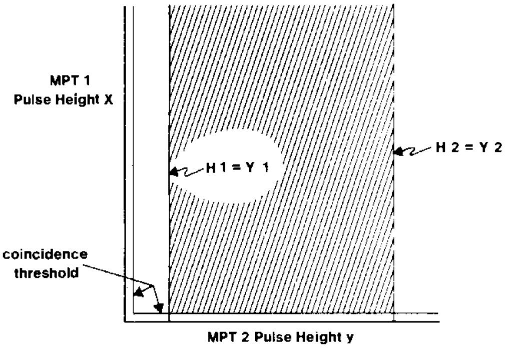

Fig. 2. Unsummed pulse height analysis, $h = y$.

analogue or digital discriminators in either a single or multichannel analyser configuration.

SINGLE-PARAMETER TRANSFORMS

Examples of three commonly used methods of pulse height analysis are depicted in Figs. 2, 3 and 4.

Figure 2 shows a 2-parameter pulse height field of 'unsummed' pulse height analysis. Phototube 1 is only used as a coincidence '*gate*'. Its pulse height is irrelevant, providing it exceeds the coincidence threshold. Events which produce a y pulse height greater than H1 but less than H2 are accepted in the pulse height window. This method of pulse height analysis was demonstrated by Hiebert and Watts,[2] and was typical of early commercial LS spectrometers.[6]

'Summed' pulse height analysis is shown in Fig. 3. In order for an event to be within the acceptance window, the sum of the x and y pulse heights must exceed level H1 and be less than level H2. Both pulse heights are linearly added prior to pulse

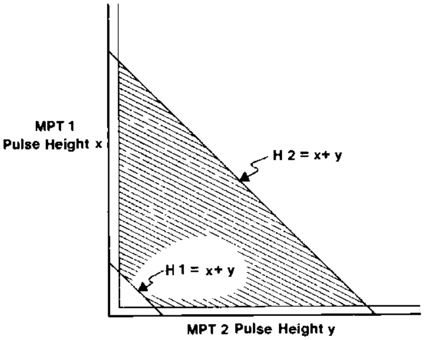

Fig. 3. Summed pulse height analysis, $h = x + y$.

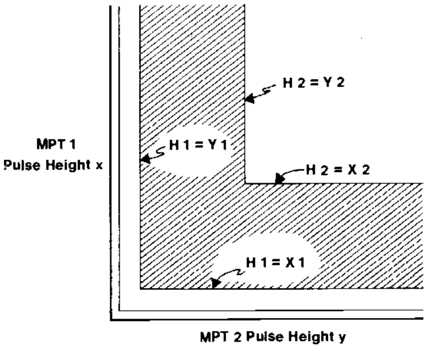

Fig. 4. Lesser pulse height analysis, $h = x \quad <y$, $h = y$ if $y \quad <x$.

height discrimination. In addition, each must exceed the coincidence threshold to be accepted. This method of pulse height analysis was demonstrated by Swank,[3] and is still used in many commercial systems.

In the 'lesser' pulse height analysis shown in Fig. 4, both x and y pulse heights must each exceed level H1 and be less than level H2 to be accepted. Thus, the lesser of the two pulse heights determines acceptance and rejection. The coincidence detector only determines the minimum level for detection. The effective coincidence threshold follows the lower level discriminator setting whenever it exceeds the threshold of the coincidence detector. This method of pulse height analysis was demonstrated by Utting,[5] and is also used in many commercial systems.

When 2-parameter information is transformed into the single parameter h, it is effectively re-mapped from a plane to a single line.[9] Thus every point on the line H1 in Figs. 2, 3 and 4 is represented by the single magnitude H1, as shown in Fig. 5. Since there are an infinite number of functions $h = f(x,y)$, a spectrum in the 2-parameter field can have many distributions in the single-parameter mapping. This may be used to explain why the pulse height spectrum of an isotope, particularly those with low energy, differs from its β-spectrum.

MATERIALS AND METHODS

The sources and instrument settings used to analyse spectra are given in Table 1. Sources contain 15 ml PPO–POPOP-toluene unless specified otherwise. Figures 6, 7 and 8 were obtained using a Searle Analytic Mark III system, while all others were with a Mark II, two Model 8928 interfaces, a Model 25609 multichannel analyser and two 100 MHz ADC's. EMI type 9750QB phototubes were used to obtain all data shown. Except for background measurements, spectral distributions are the same with RCA type 4501-V1 phototubes. Two-parameter data was read out on perforated paper tape and processed off-line. A 20-level digital grey scale conversion described by Macleod[10] was used as the log of normalised count rate, with the peak count rate being set to the maximum density. This method provides a means for obtaining both graphical and digital data simultaneously. The approximate normalised count rate at any point can be determined from the characters printed. Slight distortions in the visual image are caused by the use of a character font different from the one specified by Macleod.

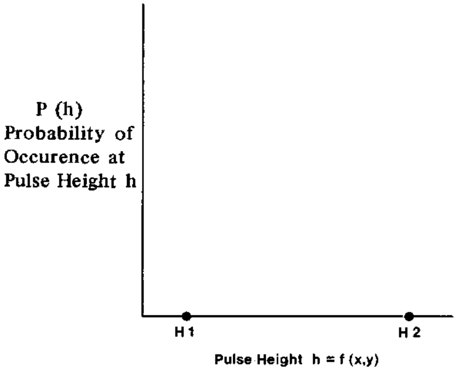

Fig. 5. Single-parameter pulse height analysis.

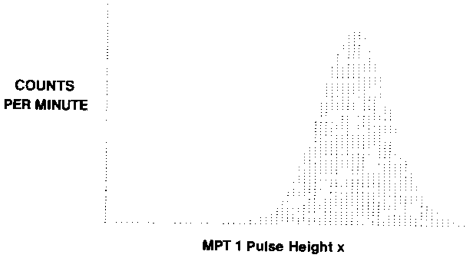

Fig. 6. Americium-241 unsummed pulse height spectrum.

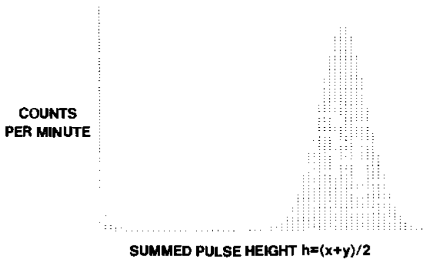

Fig. 7. Americium-241 summed pulse height spectrum.

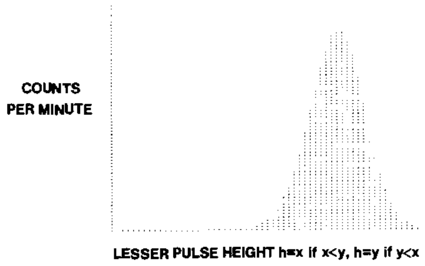

Fig. 8. Americium-241 lesser pulse height spectrum.

Table 1. Sources and instrument settings.

Figure	Source	Scintillator relative pulse height	Instrument attenuation factor
10	^{241}Am	0.7	32
6	^{241}Am	0.7	16
7	^{241}Am	0.7	16
8	^{241}Am	0.7	16
11	^{36}Cl	1.0	64
12	^{3}H	1.0	1
13	^{14}C	1.0	8
14	Blank	1.0	8
15	None	-	8
17	None	-	8
19	^{14}C	0.1	2
20	^{14}C	0.1	2
21	^{14}C	0.2	8
22	^{14}C	0.2	8
18	Chemi.	-	1

ANALYSIS OF ISOTOPE SPECTRA

The spectrum of ^{241}Am is shown 3-dimensionally in Fig. 9. The α-peak is clearly visible in the centre. This method of visualisation is acceptable for well-defined α-peaks, but is poor for less pronounced β-emitters. Americium-241 is shown as a 2-parameter density gradient in Fig. 10 using the method of Macleod.[10] Correlation of the magnitudes from each detector is indicated by conformance to a 45° line. Deviation from the 45° line is caused by non-uniform sample geometry and cathode sensitivity, as well as by the statistics of photon collection and electron multiplication.

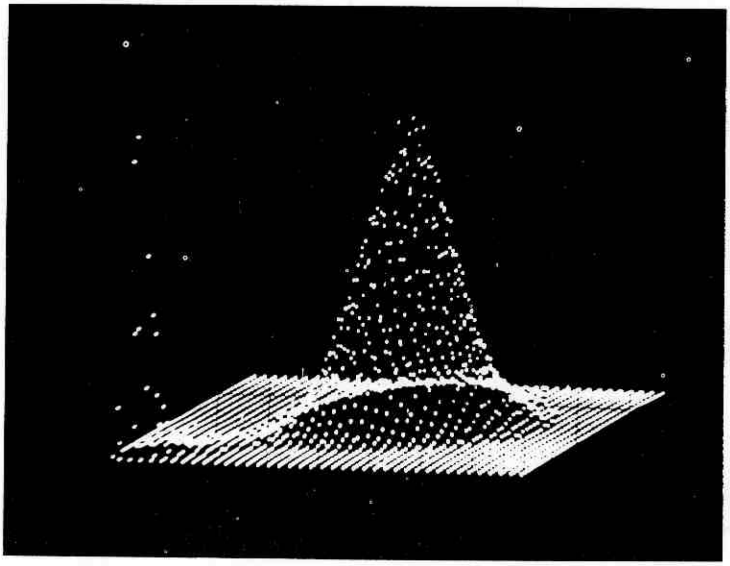

Fig. 9. Three-dimensional spectrum of ^{241}Am.

Applying unsummed, summed and lesser transforms to ^{241}Am results in the single-parameter spectra shown in Figures 6, 7 and 8. The analyser was set to stop at the same peak channel counts for all three spectra. The unsummed spectrum is broader and has poorer resolution than either the summed or lesser spectra. The summed spectrum has slightly better resolution than the lesser spectrum.

Chlorine-36, ^{3}H and ^{14}C spectra are shown in Figs. 11, 12 and 13 respectively. Because ^{36}Cl produces more photons per event, it has the best resolution as measured by its close proximity to a 45° line. In contrast, ^{3}H has poor correlation between the pulses from each phototube due to poor collection statistics associated with the small numbers of photons generated by the weak β-emitter. Thus, the greater the pulse amplitude, the greater the potential energy resolution.

When ^{14}C is quenched to 60% efficiency, the 2-parameter spectral shape nearly coincides with that of unquenched ^{3}H. In the absence of colour quenching, spectral resolution increases as the number of photons detected increases. Thus, the spectral resolution of a quenched sample containing a high-energy emitter can be poorer than for an unquenched but lower energy isotope.

ANALYSIS OF BACKGROUND SPECTRA

Blank background and empty chamber spectra are compared with a ^{14}C spectrum at the same amplification in Figs. 14, 15, and 13 respectively.

The blank background spectrum can be analysed in three parts. The high intensity region near the origin is primarily due to radioactive contamination in the glass vial. The region of intensity along the 45° line is primarily caused by Compton interactions in the liquid scintillator from external environmental and cosmic radiation. The region of intensity along the axes results from light emissions from the phototubes.

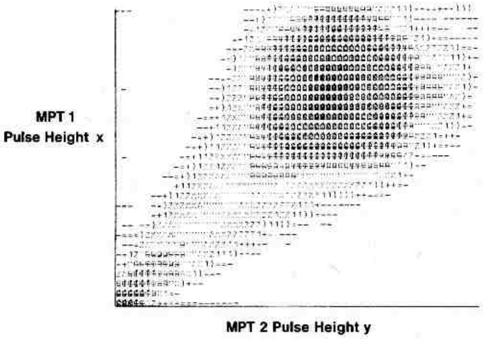

Fig. 10.

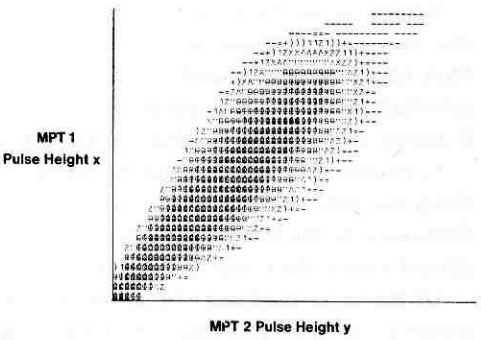

Fig. 11.

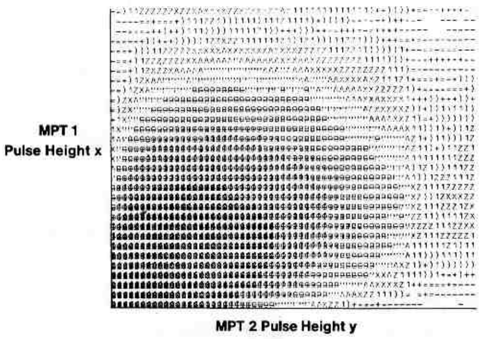

Fig. 12.

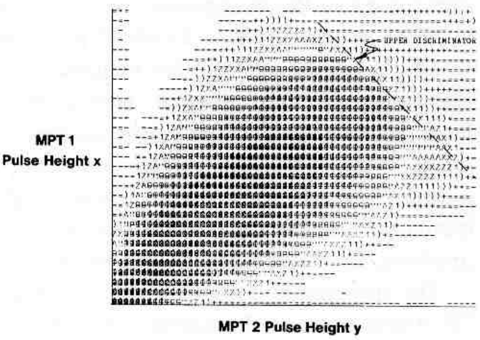

Fig. 13.

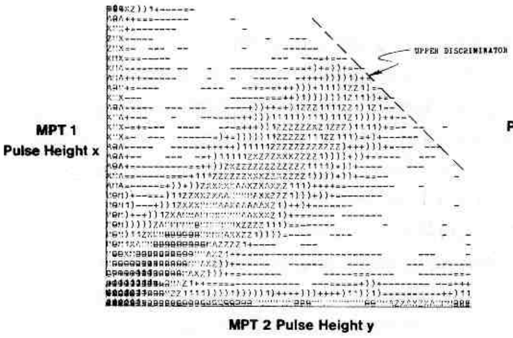

Fig. 14.

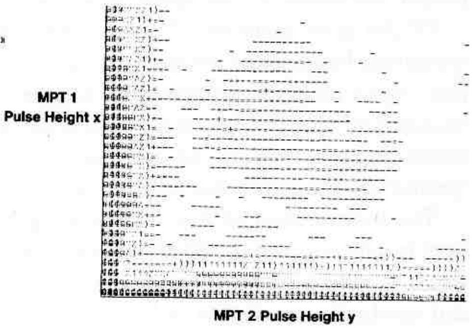

Fig. 15.

Fig. 10. Two-parameter spectrum of ^{241}Am.
Fig. 11. Two-parameter spectrum of ^{36}Cl.
Fig. 12. Two-parameter spectrum of ^{3}H.
Fig. 13. Two-parameter spectrum of ^{14}C.
Fig. 14. Background 2-parameter spectrum.
Fig. 15. Empty chamber 2-parameter spectrum.

Removing the background sample from the chamber virtually eliminates the first two sources of background (Fig. 15). The circle of weak intensity in the centre of Fig. 15 is believed to be caused by scintillations in the phototube face plates. Except for scintillations in the glass caused by external radiation, the empty chamber background is almost entirely from within the phototubes.

Comparison of the ^{14}C and background spectral distributions clearly suggests areas along the axis which can be rejected with negligible loss in ^{14}C detection efficiency. Placement of the lower level discriminator has much more control over rejecting background events than does the placement of the upper level discriminator.

Of the three methods of pulse height analysis described, the lesser method is superior for separating low pulse height background from ^{14}C events. An unsummed lower level discriminator will only reject events along one axis. A summed lower level discriminator only rejects events near the origin. A lesser lower level can be set to reject most of the background along both axes, including the region near the origin.

Since the coincidence threshold is a lesser discriminator, raising the coincidence threshold is very effective for removing background from a ^{14}C counting window. Decreasing the high voltage to both phototubes effectively raises all discrimination levels relative to the pulse height spectrum. This explains why decreasing the high voltage reduces the background more than raising the lower level discriminator of a summed instrument. With lesser pulse height analysis, raising the lower level discriminator produces the same reduction in background as decreasing the high voltage provided the phototubes are not overstressed in the former instance.

The situation is quite different for assaying ^{3}H. Because a significant number of ^{3}H events occur at the lowest detectable level near the origin, increasing the lower level discriminator significantly reduces the detection efficiency and overall sensitivity. Consequently, background events are best eliminated from a ^{3}H counting window by proper placement of the upper level discriminator.

Of the three methods of pulse height analysis, the summed method is superior for separating background from ^{3}H events. An unsummed upper level discriminator will only reject events along one axis. A lesser upper level discriminator does not reject the background along either axis. A summed upper level discriminator rejects most of the events along both axes. An even better upper level discriminator is defined by the 'greater' of the two pulse heights, as demonstrated by Hiebert and Hayes.[4]

The best method of discriminating against the background contributions along the axes is with a crosstalk discriminator.[7] A separate system is used to reject these events which is independent of the method of pulse height analysis. Consequently, efficiency and sensitivity are similar with either of the three methods of pulse height analysis.

A new technique is used to analyse the sources of light emitted from the phototubes. Since the photocathodes are designed to absorb most of the photons impinging on the surface, it is clear that only a fraction of them will pass through. Both photocathodes, therefore, form an optical attenuator to the light generated in one phototube as seen by the other. Consequently, the pulse height distribution between the two phototubes is highly unbalanced when light is generated within one of the phototubes. The 2-parameter spectrum of the empty chamber count rate shows this high concentration of events along the axes.

Recognising this uneven distribution provides a mechanism for determining which phototube produced the scintillation. In Fig. 16, the x-y plane is divided into two regions on each side of a 45° line. Events occurring in Region 1 have a larger pulse amplitude from phototube 1 than from phototube 2, and are thus assumed to originate

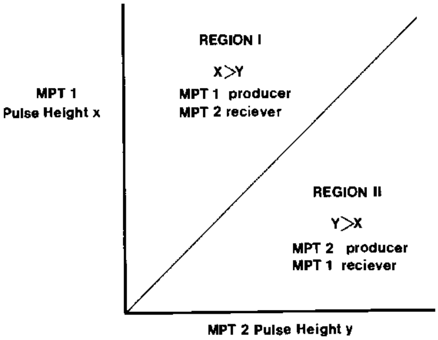

Fig. 16. Transformations for analysis of MPT light emissions.

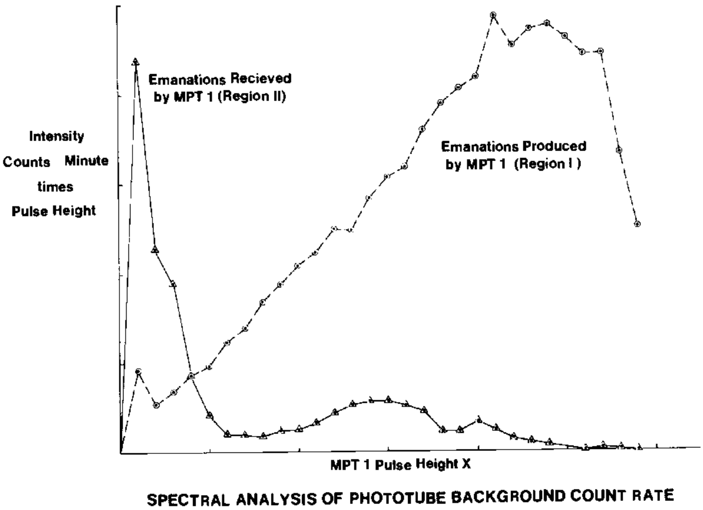

Fig. 17. Spectral analysis of phototube background count rate.

from phototube 1. Similarly, events recorded in Region II are assumed to originate in phototube 2. By adding all the events in Region I at each pulse height x, the spectrum of the emissions produced by phototube 1 can be constructed. Adding all the events in Region II at each pulse height x constructs the spectrum of emissions received by phototube 1 from phototube 2. The spectrum produced and received by phototube 1 is shown in Fig. 17. Similar integrations in the y direction will construct the spectrum of emissions which phototube 2 generated, and which phototube 1 detected from phototube 2.

Chemiluminescence and phosphorescence are other sources of background count rate. The accidental coincidence spectrum characteristic of excessive monophotonic events is shown in Fig. 18. The single photon pulse height can be determined from this spectrum. Events along the axes are probably artifacts caused by pulse pile-up in the 100 MHz ADC's used to digitise the phototube signals.

ANALYSIS OF QUENCHING

Chemically quenched and colour-quenched samples containing ^{14}C are compared in Figs. 19 and 20. Both samples have the same total counting efficiency. Events from the colour-quenched sample have a larger deviation from the 45° line than for events from the chemically quenched sample. More events have a large pulse height difference because the sample itself is an optical attenuator to light. Consequently, scintillations produced near one phototube will produce a strong signal from that phototube and a weak signal from the far phototube.[11,12]

The uneven distribution of light to each of the phototubes produces spectral distortions which can cause errors in the estimation of efficiency.[13] The lesser method of pulse height analysis has been shown to produce significantly smaller errors in efficiency than summing, as determined by the ratio method of quench correction.[14] This is because the coincidence detection efficiency is most influenced by the phototube which receives the least amount of light.[15] With summing, the signal from the phototube which receives the most amount of light contributes proportionately more to the spectra as colour quenching increases. Thus, efficiency determined from a summed pulse height spectrum is overestimated in the presence of colour quenchers.

Two samples containing urine in different surfactant cocktails are compared (Figs. 21 and 22). Using 2-parameter pulse height analysis, we can see that the Triton X-100 sample exhibits strong colour quenching even though both samples visually appear identical. Colour quenching is even visible in the 2-parameter spectrum of PCS. This spectrum would be more rounded if only chemical quenching were present.

CONCLUSIONS

Two-parameter pulse height analysis is a useful technique for analysing the signals from a liquid scintillation detector. Data presented in an array of α-numeric characters provide both digital quantitation and a graphical density gradient.

Background events generated by Compton interactions in the sample solution, radioactive contamination in the vial walls, scintillations from within the phototubes and luminescence are more easily distinguishable in the 2-parameter plane. Phototube crosstalk can be identified and electronically discriminated from scintillations within the sample.

Alternative methods of combining the signals from both phototubes prior to single-parameter pulse height analysis are readily interpreted from a 2-parameter spectrum. Spectral distribution as a function of isotope quencher and detector response can be analysed. Chemically quenched samples can be distinguished from colour-quenched samples. Although a summed spectrum has slightly better pulse height resolution than the lesser spectrum, the latter is more accurate for efficiency determinations, particularly when colour quenchers are present.

ACKNOWLEDGEMENT

The author wishes to thank Mr. Harry Engberg for performing the measurements, and Mr. J. Marshall Dudley for programming and processing the 2-parameter spectra.

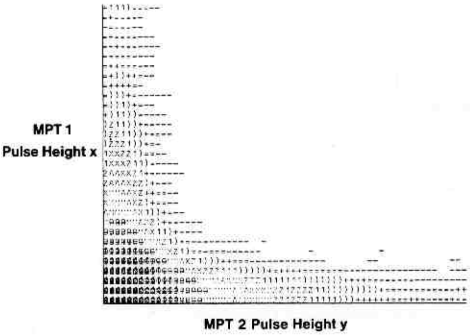

Fig. 18.

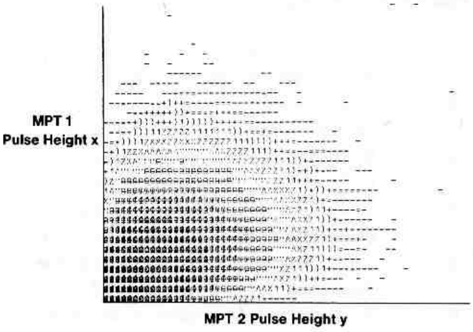

Fig. 19.

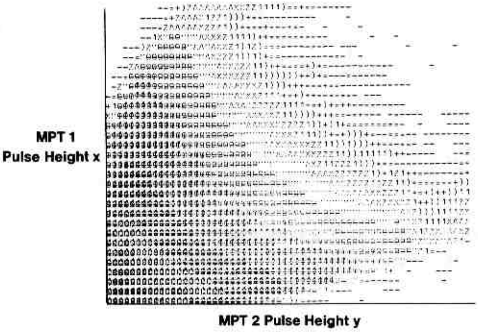

Fig. 20.

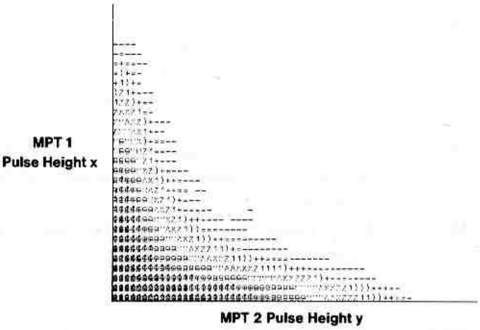

Fig. 21.

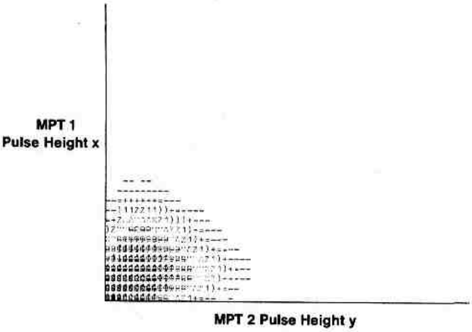

Fig. 22.

Fig. 18. Chemiluminescence 2-parameter spectrum.
Fig. 19. Carbon-14 chemically quenched to 70% efficiency.
Fig. 20. Carbon-14 colour quenched to 70% efficiency.
Fig. 21. Carbon-14 urine in Triton X-100/toluene 1/2.
Fig. 22. Carbon-14 urine in PCS.

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DISCUSSION

F. E. L. ten Haaf: Have you done a spectrum analysis on acetone as a colour quencher?

B. H. Laney: No.

J. B. Birks: In my paper I discussed two types of impurity quenching; that due to electron capture and that due to exciplex formation by excited solvent or solute molecules. There is a further type of impurity quenching, known as static quenching (the other type is dynamic quenching) which is due to the formation of donor-acceptor (charge-transfer) complexes between the quencher and solvent or solute molecules in the unexcited ground-state. Such complexes are readily formed by impurity molecules which have a high electron affinity. The complexes commonly have a charge-transfer absorption band at longer wavelengths than either of the molecular components, which extends over the solute fluorescence spectrum. They thus function both as an impurity quencher and a colour quencher. Further details will be found in my book *Photophysics of Aromatic Molecules* (Wiley-Interscience, London and New York, 1970).