

Chapter 3

Colour Quenching in Liquid Scintillation Coincidence Counters

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INTRODUCTION

The liquid scintillation counting technique requires the addition of a radioactive sample to a solution of scintillating material prior to measurement. Usually the scintillation properties of the solution are deteriorated due to the addition of the sample and this effect is called *quenching*. *Chemical quenching* is a common name for the phenomena that reduce the number of photons per MeV primary particle energy, initially produced in a scintillation event. The term *colour quenching* or *optical quenching* is used to denote the subsequent loss of photons along their paths to the photocathodes of the photomultiplier tubes in the detector.

Various authors have discussed the experimental aspects of quenching and, in particular, the difficulties which are sometimes encountered in the determination of the counting efficiency of coloured samples.¹⁻³ Instrumental methods for determining the efficiency often require different calibration curves for samples of different composition and this seems to imply that the spectral response of the instrument in each case is affected in a different way.

So far, however, little effort has been made to explain these effects in terms of spectrometer response. No doubt the reason for this is the extreme complexity of the problem, where many parameters are involved.

In 1969 Neary and Budd⁴ published pulse height analyser spectra obtained from liquid scintillation samples, quenched by different chemicals. They showed that coloured samples produce broader pulse height distributions than uncoloured samples with approximately the same counting efficiency. The authors explain this by suggesting that the light produced in different parts of a sample will take paths with different lengths. For coloured samples this results in a pulse height variance which adds to the normal statistical variance of a scintillation event.

In 1971 Kaczmarczyk⁵ discussed a detailed model of the response of a liquid scintillation counter which, however, did not take colour quenching into consideration.

In this chapter a simple mathematical model is set up, which illustrates qualitatively some typical features of chemical and optical quenching. Pulse height spectra generated by the model for different conditions will be compared with experimental data.

A SIMPLE MATHEMATICAL MODEL FOR QUENCHING

Assuming a linear relationship between pulse height and energy, we represent the height $h(E)$ of a pulse caused by a β -particle with the energy E , in the absence of colour quenching as:

$$h(E) = qcE \quad (1)$$

where c is the conversion factor for an unquenched sample and q represents the pulse height reduction factor due to chemical quenching.

The range of low energy β -rays can be considered small in comparison with the sample dimensions. In this case we may assume that the initial absorption of each single β -particle, the transfer of part of its energy to scintillator molecules, and the subsequent conversion of the energy into light, all occur in a small volume element within the sample. By definition the effect of chemical quenching is confined to the same small volume element which, for simplicity, we shall regard as a point.

In the presence of colour quenching, the additional attenuation of the pulse due to light absorption has to be accounted for (see Fig. 1(a)). The position of each scintillation event must now be considered. However, to account for all the parameters involved would lead to a complicated multiple integral, but we simplify matters by considering the system as if it were one-dimensional. In the original sample (Fig. 1(a)), the activity is spread evenly throughout the three-dimensional sample. In the mathematical model the 'sample' is represented by the line joining the photomultiplier photocathodes, which has a length $2d$ (see Fig. 1(b)). The activity is considered to be evenly spread along this line segment.

We now consider one scintillation event with a position given by a point on the line segment with the coordinate x , where $-d > x > d$. There are now only two possible light paths: one with a length $d + x$, towards photomultiplier 1; the other, with a length $d - x$, towards photomultiplier 2. If the optical attenuation constant is μ , the light traversing the first path will be attenuated by a factor $\exp(-\mu(d + x))$. The light traversing the second path will be attenuated by a factor of $\exp(-\mu(d - x))$. Consequently, the pulse heights h_1 and h_2 at the outputs of the photomultipliers will be:

$$h_1(E, x) = \frac{1}{2} \exp(-\mu(d + x)) [qcE] \quad (1a)$$

and

$$h_2(E, x) = \frac{1}{2} \exp(-\mu(d - x)) [qcE] \quad (1b)$$

In coincidence counters it is customary to add these two pulses together in a pulse summation circuit (Fig. 6) prior to analysis. The pulse height at the output of this circuit is then given by:

$$h_s(E, x) = h_1 + h_2 = \frac{\exp(-\mu(x - d)) + \exp(-\mu(x + d))}{2} [qcE]$$

This can also be written as:

$$h_s(E, x) = \cosh(\mu x) [\exp(-\mu d) qcE] \quad (2)$$

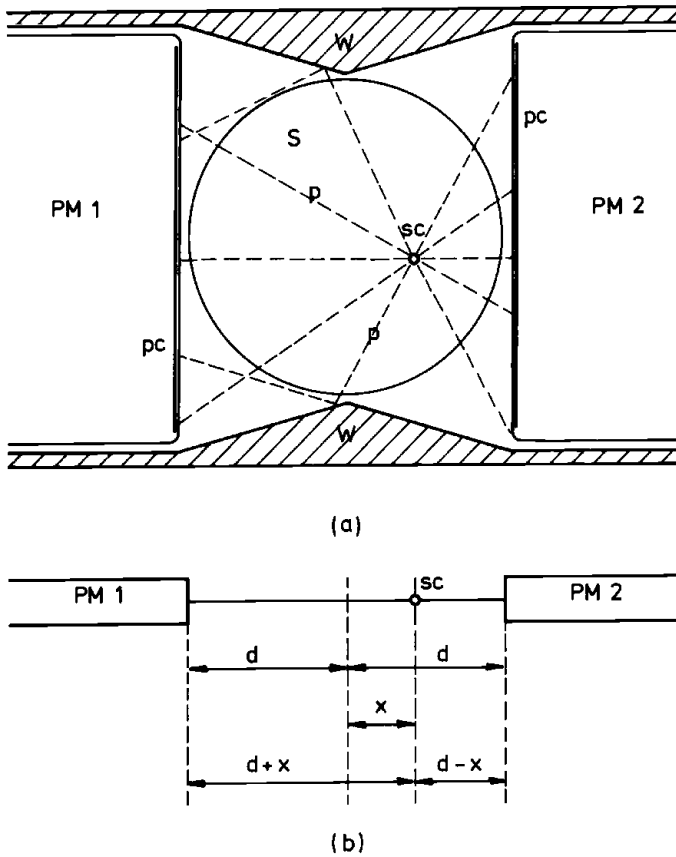


Fig. 1. (a) Schematic cross-section through liquid scintillation detector chamber with photomultipliers (PM) and their photocathodes (pc), reflective chamber walls (W), sample (S), arbitrary scintillation event (sc) and photon paths (p). (b) Simplified one-dimensional representation as discussed in text.

Equation (2) shows that, unless $\mu = 0$, the pulse height for a given energy depends on the position of the scintillation event in the sample.

The functions h_1 , h_2 and h_s are represented graphically in Fig. 2 for arbitrary values of E , q and μ . This figure shows that a pulse from an event at x_1 will be counted in a coincidence system, but a larger pulse at x_2 will not be counted. The reason is that in the first case h_1 and h_2 are both above the one photoelectron level, a condition which is required to register the event as a coincidence. In the second case h_1 is below the one photoelectron level, and a coincidence becomes improbable. In fact, there is a low probability that any events will be counted which occur outside the region shown shaded in Fig. 2. This region will be smaller for larger values of μ , and consequently the coincidence losses will increase. If μ is small and E is sufficiently large, the coincidence losses will be insignificant.

It is possible to derive an analytical expression for the single energy response of the model (see also Fig. 3). For this purpose assume that a number of scintillation events, equal to the unit of activity, are distributed evenly in the 'sample', that is, along the line

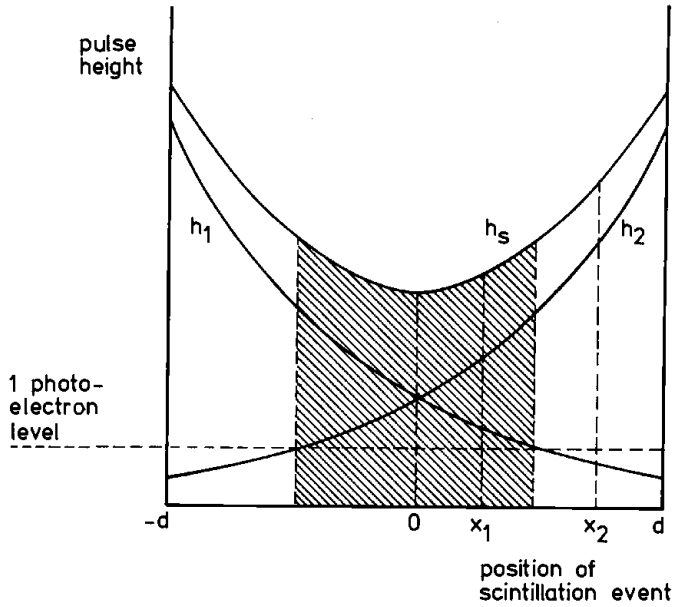


Fig. 2. Graphical presentation of heights of single pulses and summed pulse as a function of x for arbitrary fixed values of E , q and μ .

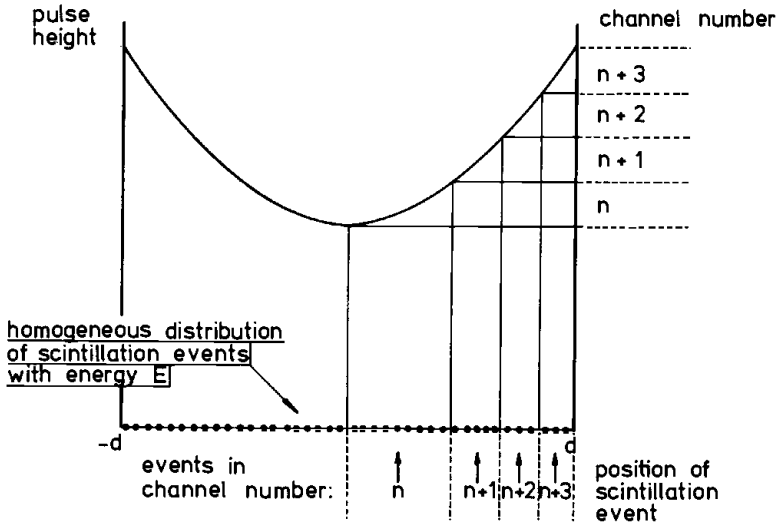


Fig. 3. Illustration of the correlation between the position of events with the same energy in a sample and the channel in which they will be counted, if optical quenching is present.

segment $(-d, d)$. All events have the same energy E . If we call the number of events n , we have:

$$\int_{-d}^{+d} dn = 1, \text{ while } \int_{-d}^{+d} dx = 2d$$

Consequently:

$$dx = 2d \cdot dn \quad (3)$$

From Eqn. (2) we get, by differentiation with respect to x :

$$\left(\frac{dh_s}{dx}\right)_{E,x} = \mu \sinh(\mu x) \cdot h_s(E, 0) \quad (4)$$

where $h_s(E, 0)$ stands for $\exp(-\mu d)qcE$, which is the pulse height for an event in the centre of the sample. After inverting Eqn. (4) and substituting Eqn. (3), we have:

$$\left(\frac{dn}{dh_s}\right)_{E,x} = \frac{1}{2d \mu \sinh(\mu x) h_s(E, 0)} \quad (5)$$

Equation (5) represents the pulse height response for a sample with a homogeneous distribution of scintillation events, all having the same energy E . In Fig. 4 two examples are shown. Figure 4(a) represents the effect of pure chemical quenching, where there is no dispersion at all. This is, of course, due to the fact that the model does not account for statistical pulse height deviations. In reality some dispersion does occur, especially at small pulse heights. Figure 4(b) shows the effect of pure optical quenching. There is appreciable pulse height dispersion and we note that, at the high energy side, there is a region where pulses will be lost if a coincidence condition is imposed, as discussed previously.

In order to construct the pulse height response of the model for a given β -emitter we need an expression for the β -energy distribution. Nuclear theory provides a distribution function which, after some remodelling takes the form:

$$p(E) = kF(E + 511)(E_{\max} - E)^2 (E^2 + 1022E)^{1/2} \quad (6)$$

where $p(E)$ is the probability that a β -particle has an energy E (in keV). The function F is a correction factor for the influence of the electrical field of the nucleus and depends on E and the nuclear charge Z . For simplicity we will disregard this effect and make $F = 1$. The constant k will serve to normalize the distribution function in such a way that:

$$\int_0^{E_{\max}} p(E) dE = 1$$

From Eqn. (2) we have:

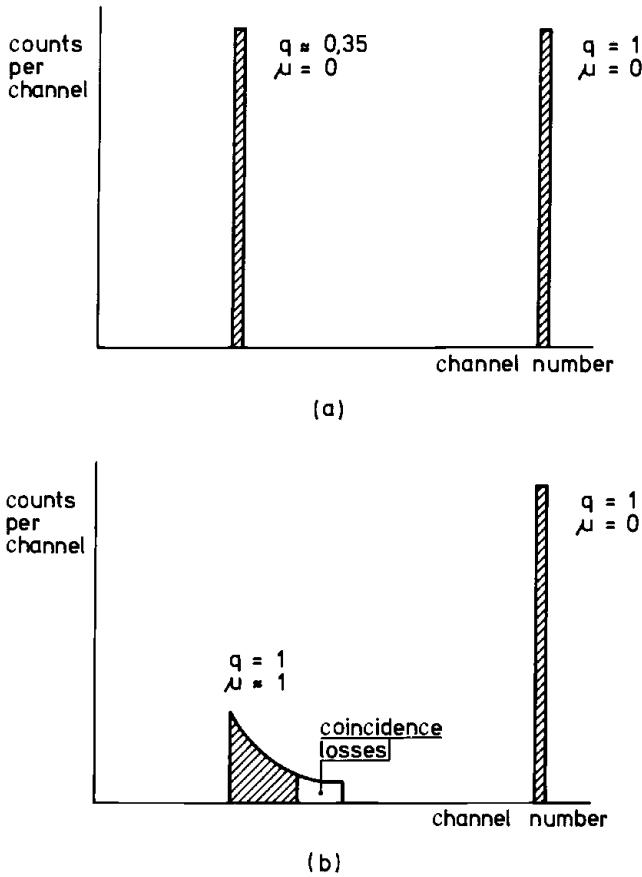


Fig. 4. Single energy response from a chemically quenched sample (a) and an optically quenched sample (b) according to the model.

$$E = \frac{h_s}{\cosh(\mu x) \exp(-\mu d) q c} \quad (7)$$

Assuming values for E_{\max} , c , q and μ , one can now find the pulse height distribution $p(h_s)$ for any given value of x , by substituting Eqn. (7) in Eqn. (6) and calculating the value of Eqn. (6) for a sufficiently large number of values of h_s . To find the pulse height response for an evenly distributed activity one has to repeat this procedure for a sufficiently large number of equidistant x values, and to superimpose all the obtained distributions.

Figure 5 shows some results, in the form of integral pulse height distributions, obtained by computer calculation, for different values of q and m , where $m = \mu/\log 2$. As E_{\max} was given a value of 158 keV, the spectra can be associated with carbon-14. The constant c was taken as 0.7 photoelectrons/keV, and the distance d as 1 cm.

To obtain pulse height distributions representative of a coincidence counter, the condition that both h_1 and h_2 [Eqns. (1a) and (1b)] have to be at least equal to one for an event in order to be counted, was 'built into' the computer program.

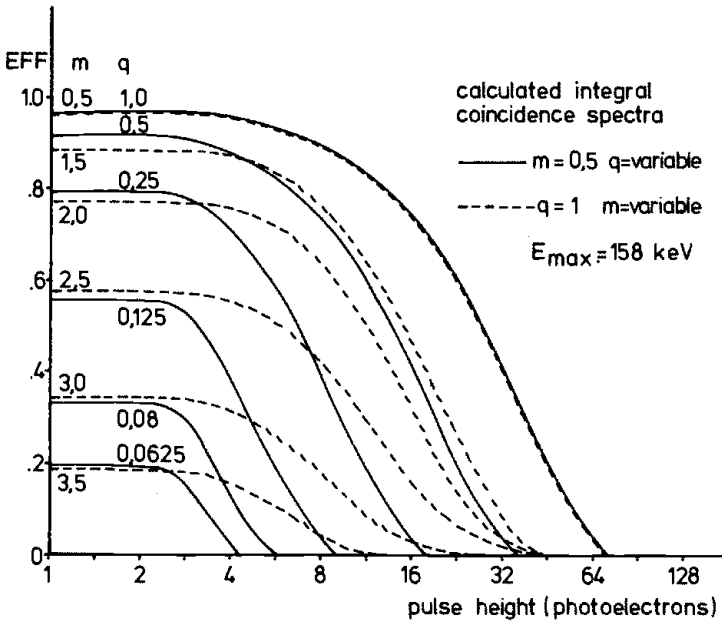


Fig. 5. Pulse height distribution curves, calculated by computer, for values of q and m . ($E_{max} = 158$).

The pulse height distributions shown in Fig. 5 consist of two series. The full lines represent distributions for different values of q , simulating different degrees of chemical quenching. The parameter m here has a constant low value of 0.5. The reason for making it 0.5, and not 0, is a trick to account for the appreciable geometrical effect in a coincidence counter.

The dotted lines represent pulse height distributions for different values of m , which simulate different degrees of optical quenching. Here q is constant and has the value of 1.

A comparison of the two series shows that the pulse height distributions associated with optical quenching cover a much wider energy range than distributions associated with chemical quenching.

Figure 9 shows calculated integral pulse height spectra with and without the coincidence condition imposed. It demonstrates that optical quenching is associated with large coincidence losses, whereas in the case of chemical quenching the coincidence losses are slight.

COMPARISON WITH EXPERIMENTAL RESULTS

Experimental pulse height distributions were obtained by means of a commercial liquid scintillation counter (Philips model PW 4510). This instrument has pulse-amplifiers of the linear type, but it is provided with a logarithmically calibrated gain control. Spectra were taken in the integral mode with a fixed lower level setting, and the logarithm of the gain was varied in steps.

Under normal conditions coincidence spectra are measured, but if the coincidence

circuit output cable is disconnected from the coincidence gate (see Fig. 6), the gate remains open and single spectra can be obtained.

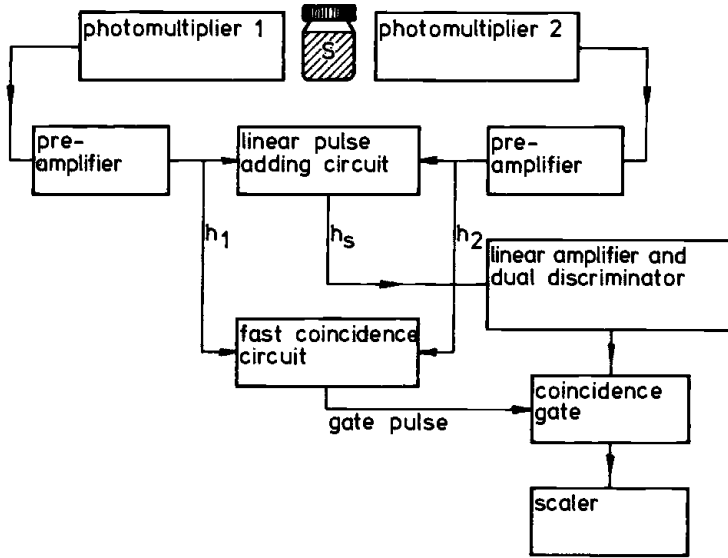


Fig. 6. Simplified block diagram of liquid scintillation counter.

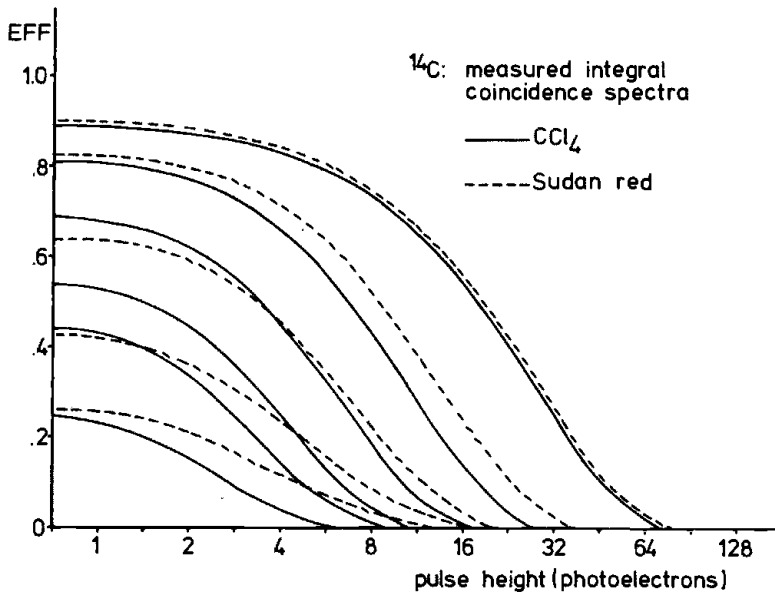


Fig. 7. Coincidence spectra of chemically quenched and optically quenched samples of carbon-14.

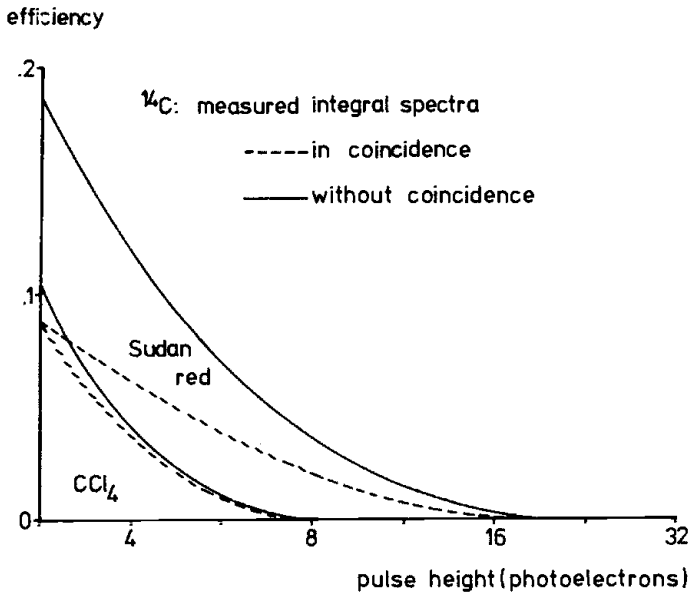


Fig. 8. Coincidence and single spectra of a chemically quenched and an optically quenched sample. The non-coincident spectra could be measured down to the three photoelectron level without disturbance from the thermal noise of the photomultipliers.

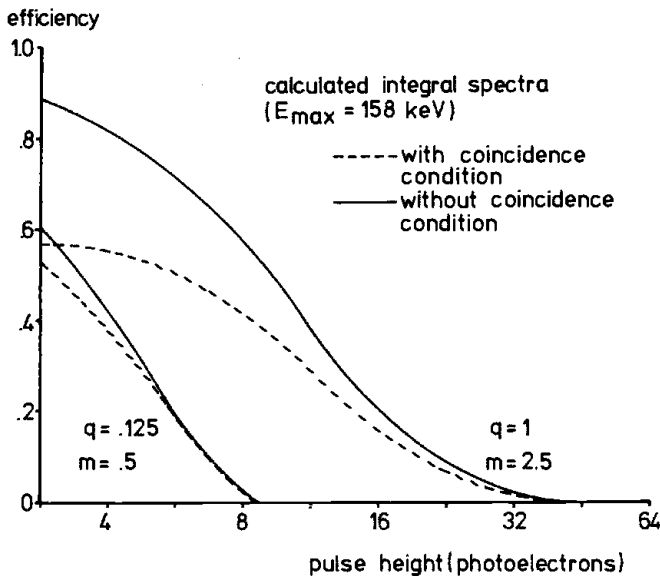


Fig. 9. Calculated integral pulse height spectra, with and without the coincidence condition imposed.

Two series of ^{14}C -spiked samples were measured, all having the same activity and scintillator composition. The scintillator was butyl-PBD (6 g/l toluene). To one series increasing quantities of CCl_4 had been added and to the other increasing quantities of Methyl-red. In Fig. 7 the coincidence spectra obtained from these samples are shown, where the count rate is divided by the sample activity. On the horizontal axis the pulse height is represented by the estimated equivalent number of photoelectrons.

The colour-quenched samples, when compared with chemically quenched samples of approximately equal integral counting efficiency, have the same tendency towards broader pulse height distributions as shown by the calculated spectra in Fig. 5.

Figure 8 represents the coincidence and single spectra of a colour-quenched sample and a chemically quenched sample, having approximately the same integral counting efficiency in coincidence. The figure shows that the colour-quenched sample suffers much larger coincidence losses than the chemically quenched sample. This is in agreement with the calculated results shown in Fig. 9.

CONCLUSION

Colour-quenched liquid scintillation samples show broader energy distributions and larger coincidence losses than chemically quenched samples which have approximately the same counting efficiency. These effects can be explained qualitatively by means of a simple mathematical model.

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DISCUSSION

J. B. Birks: Some instrument manufacturers have claimed that quench correction methods using channel ratios and/or external standards are applicable to both chemical quenching and colour quenching. The present analysis shows that this is not valid. An internal standard appears necessary to correct properly for colour quenching.

F. E. L. ten Haaf: I quite agree that this is true for the general case where both chemical and optical quenching vary strongly within a given series of samples. However, if the quenching is mainly due to one or the other type, the external standard and/or channels ratio method can be used successfully, provided of course the proper calibration curve is used for each type. At very low degrees of quenching one might use one and the same curve without making large errors.

J. A. B. Gibson: How good is the agreement between the theoretical and practical distributions?

F. E. L. ten Haaf: Not very good, since the theoretical model is a very simplified one. I look on this as a mathematical exercise, which has shown that our simplified model does bear considerable relation to reality.