Chapter 4

A Simple Mathematical Model of a Liquid Scintillation Counter

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INTRODUCTION

Although it is possible to understand each of the basic processes taking place in a liquid scintillation counter, the number of parameters involved, the complex way in which they interact with each other and the difficulty in assessing some of them quantitatively make it virtually impossible to give an accurate mathematical description of the complete liquid scintillation counting process. However, even imperfect mathematical models may eventually become helpful in understanding certain aspects of this process and possibly lead to improvements in measuring methods or instrument design. In recent years, various models have been proposed.[1-3]

The mathematical model presented in this study is a 3-dimensional extension of the model discussed earlier by one of the authors.[4] It is used to generate probability distributions representing pulse height distributions from differently quenched liquid scintillation samples. These are compared with experimentally obtained pulse height distributions. Calculated channels-ratio calibration curves, based on the pulse height distributions produced by the model, are compared with experimentally obtained channels-ratio curves.

THE MATHEMATICAL MODEL

In order to reduce the complexity of the model a number of simplifying assumptions are made. Secondary effects, such as re-emission of light following an absorption, and effects caused by the vial wall and cap are neglected. The bottom of the vial and the detector chamber are considered to be perfect mirrors. No allowance is made for the spectral distribution of light emission and absorption. The relation between pulse height and β-energy is assumed to be linear. Statistical fluctuations in any parameter concerned are not taken into account.

We now consider a scintillation event caused by a β-particle with an energy E at an arbitrarily chosen point P (ρ, α, y) in the sample, assuming that its light is emitted evenly in all directions (see Fig. 1). In the absence of quenching, we suppose that one half of the light goes to photomultiplier 1, whereas the other half is collected by

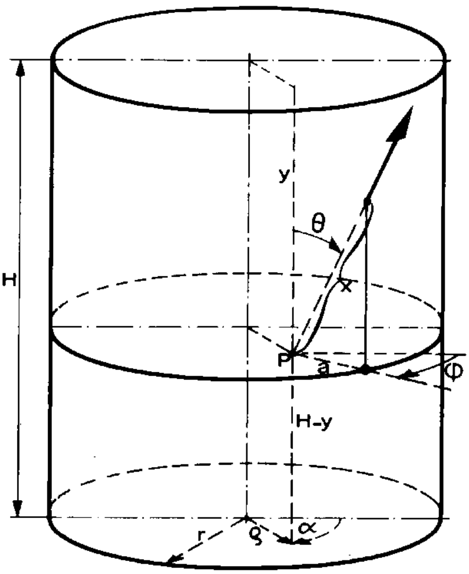


Fig. 1.

photomultiplier 2. The output pulse heights of both photomultipliers will then be equal and proportional to E:

$$h_1 = h_2 = \tfrac{1}{2} cE \qquad (1)$$

In the presence of quenching, h_1 and h_2 will generally be different:

$$h_1 = f_1 cE \qquad (2a)$$

$$h_2 = f_2 cE \qquad (2b)$$

For f_1 and f_2 we find:

$$f_1(\rho, \alpha, y) = \frac{q}{4\pi} \int_{\phi = \pi/2}^{3\pi/2} \int_{\theta = 0}^{\pi} \exp(-\mu x) \sin\theta \, d\theta d\phi \qquad (3a)$$

$$f_2(\rho, \alpha, y) = \frac{q}{4\pi} \int_{\phi = -\pi/2}^{\pi/2} \int_{\theta = 0}^{\pi} \exp(-\mu x) \sin\theta \, d\theta d\phi \qquad (3b)$$

Here q is the chemical quench factor ($0 < q \leqslant 1$), μ is the optical attenuation coefficient of the sample material ($\mu \geqslant 0$) and x is the length of the light path through the sample. The value of x is a function of the coordinates ρ, α, y, θ and ϕ, and of the height H and radius r of the sample:

$$\left.\begin{array}{ll} x = y \sec\theta & \text{for } 0 \leqslant \theta \leqslant \arctan \dfrac{a}{y} \\ x = a \operatorname{cosec}\theta & \text{for } \arctan \dfrac{a}{y} \leqslant \theta \leqslant \pi + \arctan \dfrac{a}{y-H} \\ x = (y-H)\sec\theta & \text{for } \pi + \arctan \dfrac{a}{y-H} \leqslant \theta \leqslant \pi \\ \text{where} & \\ a\sqrt{r^2 - \rho^2 \sin^2(\alpha-\phi)} - \rho\cos(\alpha-\phi) & \end{array}\right\} \quad (4)$$

If linear pulse summation is applied, the height of the pulse presented by the detector to the analyser as a result of a scintillation event at $P(\rho, \alpha, y)$ is

$$h_s(\rho, \alpha, y) = \frac{qcE}{4\pi} \int_{\phi=0}^{2\pi} \int_{\theta=0}^{\pi} \exp(-\mu x) \sin\theta \, d\theta \, d\phi \quad (5)$$

This pulse, however, will only be counted if both h_1 and h_2 are above the one-photoelectron level.

In order to find the pulse height distribution for a given β-emitter divided homogeneously in a sample with given quenching parameters, the integrations (3) and (5) have to be performed for a sufficient number of points in the sample and a representative number of values of the β-energy distribution function. We chose 4400 points, divided evenly throughout the sample and properly weighed for the volume they represented. However, the actual number of points required for calculating each pulse height distribution could be reduced to 550 on symmetry considerations. The time required by the computer to calculate one pulse height distribution was about 4 min.

For the β-energy distribution we used the expresssion

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

where $p(E)$ is the probability that a β-particle has an energy E (in keV). The function F, which is a correction factor for the influence of the electrical field of the nucleus, was disregarded and made equal to 1. The constant K was used to normalise the function in such a way that

$$\int_0^{E_{max}} p(E) dE = 1 \quad (7)$$

It is well known from practical measurements that instrumental quench correction methods, such as the channels-ratio method and the external standard method, are sensitive to the shape of the pulse height spectrum. It therefore seemed interesting to calculate channels-ratio curves from pulse height distributions generated by the model and to compare these with experimental data. In order to represent quenchers with mixed chemical and colour quenching properties in a form suitable for use in the model, we assumed

$$\left.\begin{aligned} q &= \exp(-\gamma Q) \\ \mu &= \gamma M \end{aligned}\right\} \tag{8}$$

where γ fulfils the function of a concentration.

Values for the integral efficiency and channels-ratio were obtained from five series of calculated pulse height distributions, each for a different pair of values of Q and M, according to the following table:

Curve	Q	M
A	1	0
B	0.75	0.25
C	0.5	0.5
D	0.25	0.75
E	0	1

The channels-ratio curves obtained by means of these values are shown in Fig. 2. The calculated distributions were based on a value of 158 keV for E_{max}. The curves can, consequently, be associated with ^{14}C.

A few calculated pulse height distributions are shown in Fig. 3. They are the distributions that produced the 'calibration points' indicated by the dotted circles in Fig. 2.

EXPERIMENTAL RESULTS

Experimental data were obtained by means of a commercial liquid scintillation counter (Philips PW 4540). For comparison with the calculated results we prepared several series of progressively quenched ^{14}C samples. All samples contained 50,000 disintegrations min^{-1} ^{14}C in the form of ^{14}C-hexadecane. They were quenched with CCl_4, Azobenzene and Sudan red, respectively, and of each we prepared three series with different scintillator concentrations. Figures 4 to 6 show the ratio of the integral counting efficiency of the quenched samples to that of the unquenched sample of each series.

The carbon tetrachloride shows its typical chemical quenching properties by producing markedly different results at different scintillator concentrations. Sudan red is practically insensitive to changes in the scintillator concentration, demonstrating its predominant colour quenching properties. Azobenzene seems to assume an inter-

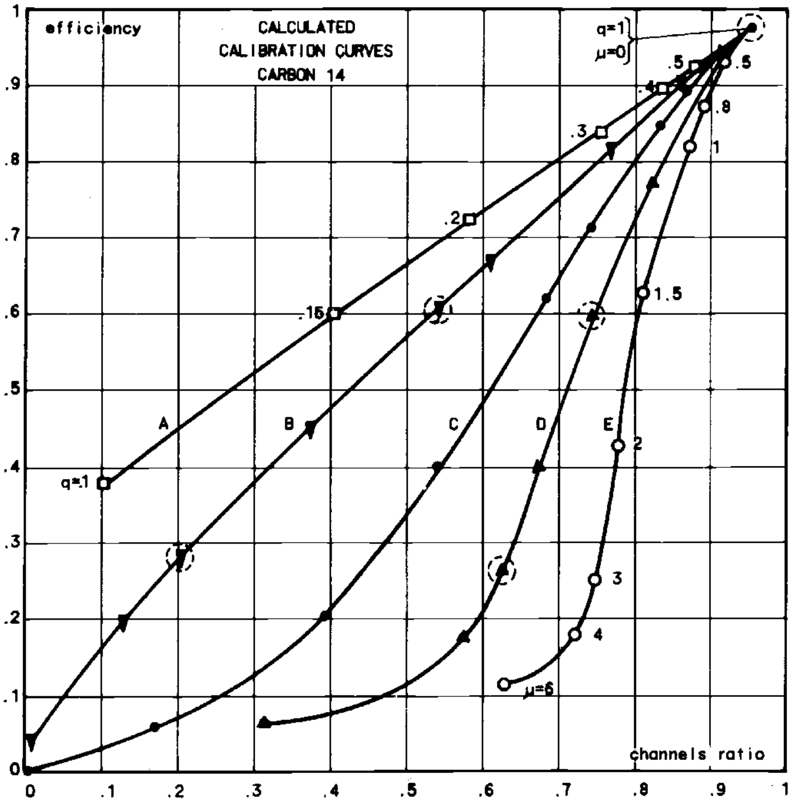

efficiency
CALCULATED
CALIBRATION CURVES
CARBON 14
q=1
μ=0
A
B
C
D
E
q=1
μ=6
channels ratio
.15
.2
.3
.4
.5
.8
1
1.5
2
3
4
.5

Fig. 2.

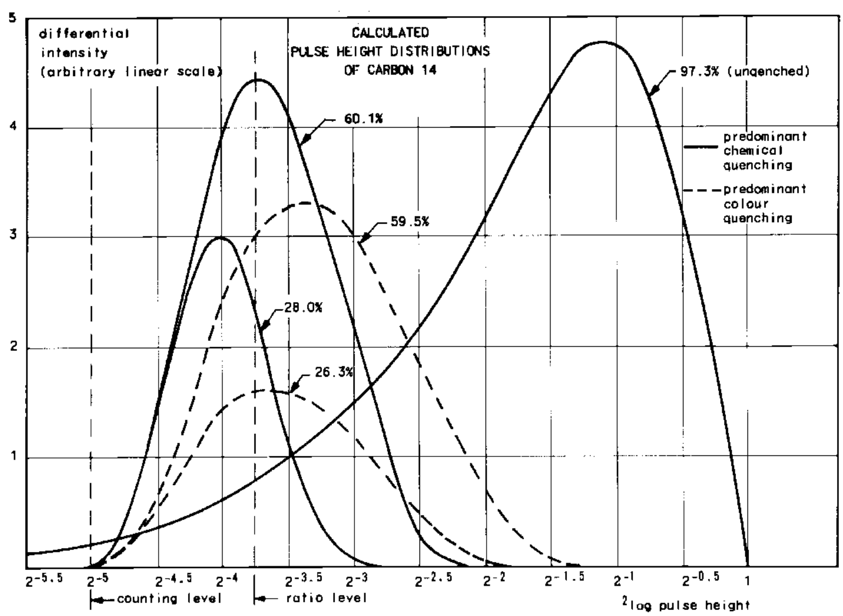

differential
intensity
(arbitrary linear scale)
CALCULATED
PULSE HEIGHT DISTRIBUTIONS
OF CARBON 14
97.3% (unqenched)
60.1%
59.5%
28.0%
26.3%
predominant
chemical
quenching
predominant
colour
quenching
counting level
ratio level
2log pulse height

Fig. 3.

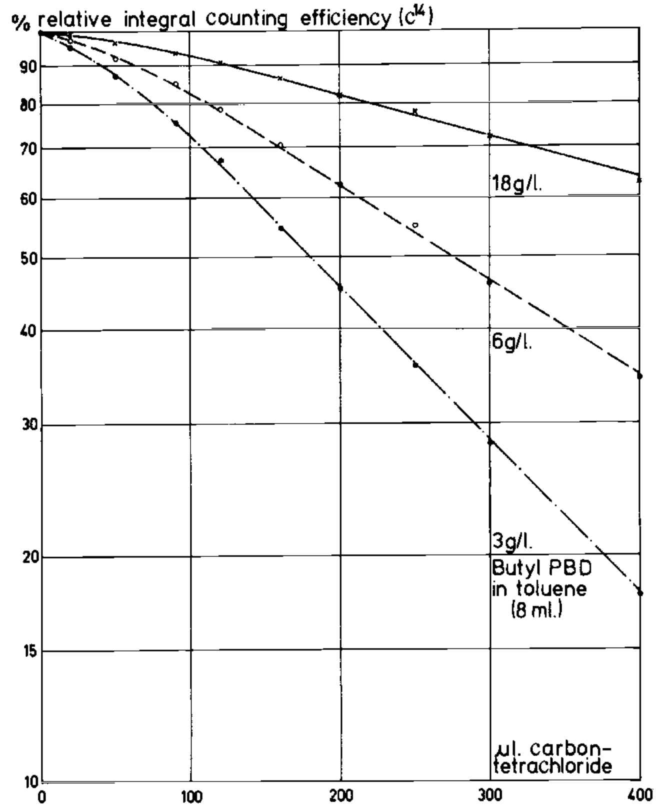


Fig. 4.

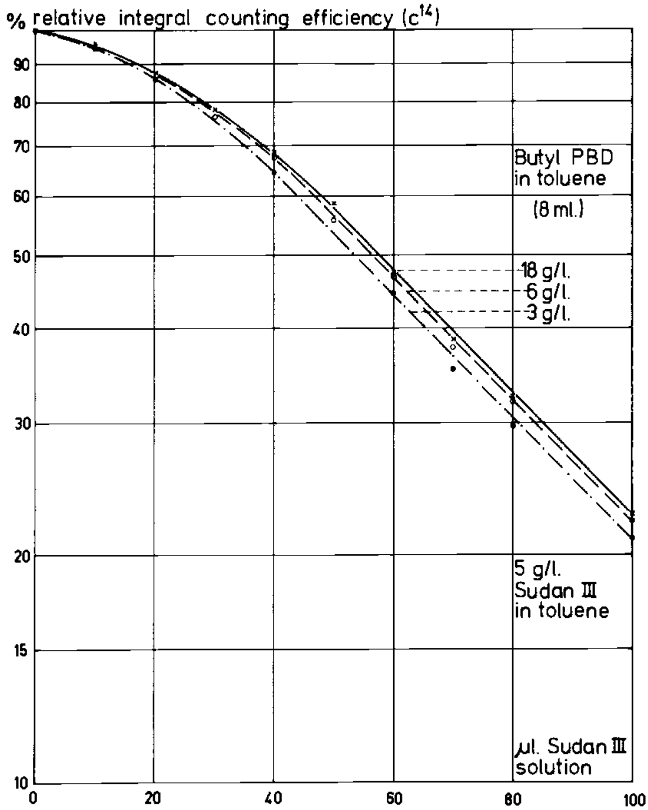
mediate position. These considerations are in agreement with Fig. 7, which shows experimental quench curves obtained with the three quenching agents previously mentioned. The scintillator was 6 g l^{-1} butyl-PBD in toluene. The curves represent integral counting efficiency versus sample channels-ratio.

Of a few samples (indicated by circles in Fig. 7) pulse height distributions were determined by means of an experimental logarithmic pulse height analyser. They are shown in Fig. 8. As each channel of this analyser represents a pulse height increment $H + \Delta H = \sqrt{2}$, the horizontal scale of Fig. 8 is equivalent to that of Fig. 3.

DISCUSSION AND CONCLUSION

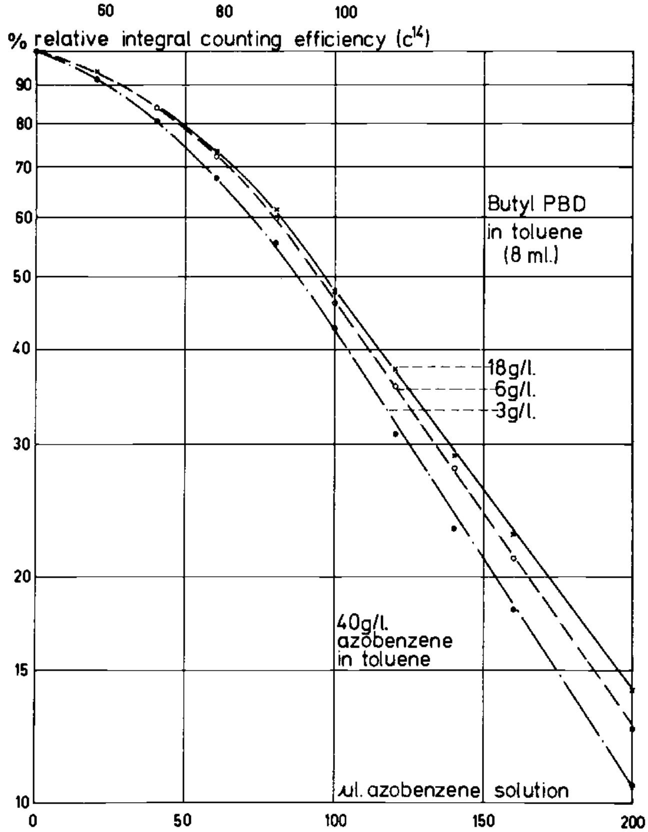
The experimental, as well as the calculated, pulse height distributions (shown in Figs. 3 and 8, respectively) show the typical differences in shape attributable to different types of quenching. It should be noted that the presentation on a logarithmic pulse height scale has two advantages. One is that it is easier to represent a wide range of pulse amplitudes in a single figure. The second advantage is that a multiplication in the pulse height direction amounts to a simple translation. This makes it easier to compare changes in spectral shape due to different quenching conditions.

When comparing the calculated with the experimental channels-ratio curves (Figs. 2 and 7), one is tempted to conclude that, apparently, carbon tetrachloride and



% relative integral counting efficiency (c14)
90
80
70
60
50
40
30
20
15
10
0
20
40
60
80
100
Butyl PBD
in toluene
(8 ml.)
18 g/l.
6 g/l.
3 g/l.
5 g/l.
Sudan III
in toluene
μl. Sudan III
solution

Fig. 5.



% relative integral counting efficiency (c14)
90
80
70
60
50
40
30
20
15
10
0
50
100
150
200
Butyl PBD
in toluene
(8 ml.)
18g/l.
6g/l.
3g/l.
40g/l.
azobenzene
in toluene
μl. azobenzene solution

Fig. 6.

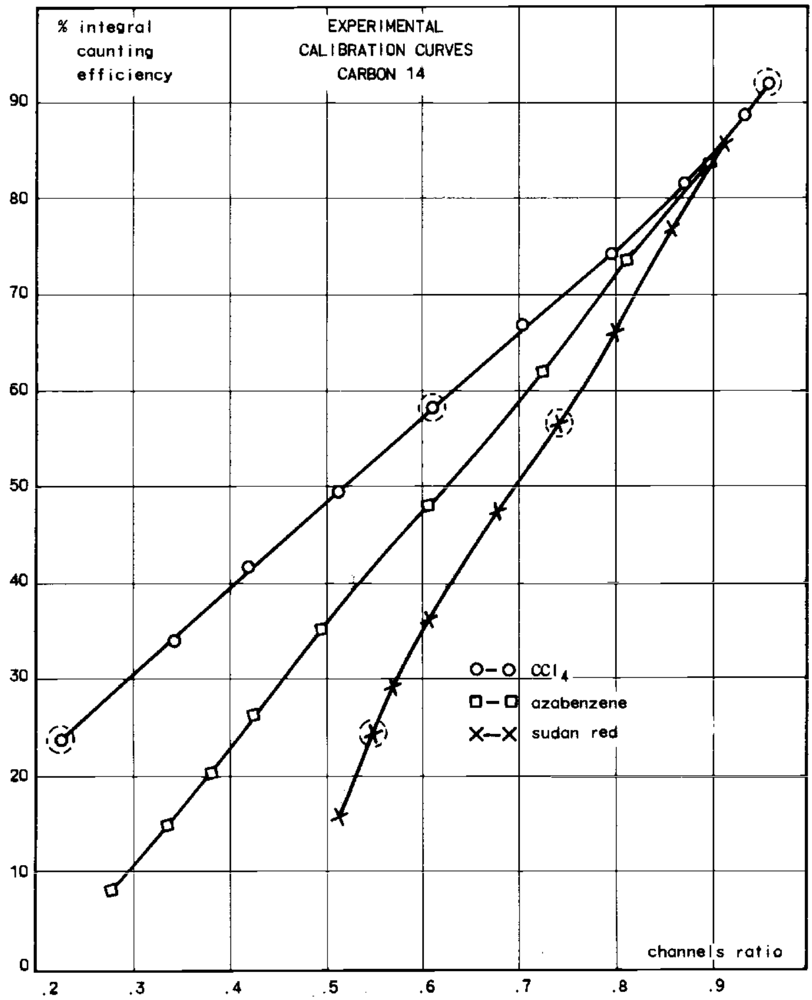


Fig. 7.

Sudan red do not represent truly extreme conditions. But the discrepancies are more than likely due to the simplifying assumptions that were made when the mathematical model was set up. However, the model correctly 'predicts' the trend found in the experimental data, where colour quenchers produce a more rapid decrease of efficiency with channels-ratio than chemical quenchers.

It should be mentioned that we purposely considered integral ^{14}C-efficiencies in order to obtain substantially different results for different types of quenching that could easily be measured and demonstrated. However, such measuring conditions would have a detrimental effect on measurement accuracy in practical liquid scintillation counting. Fortunately, this effect can be strongly reduced by using a counting window. The effects mentioned are most pronounced at low pulse heights and can be appreciably reduced by means of the lower level of the counting window.

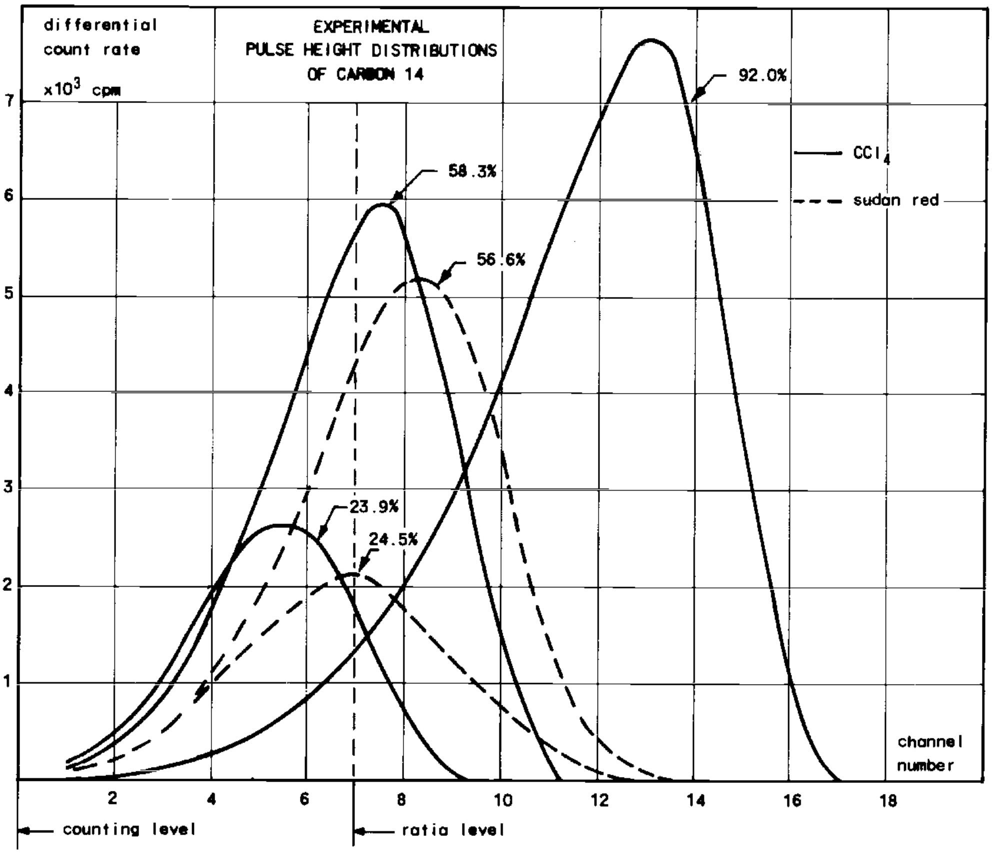


Fig. 8.

If external standardisation with a high-energy γ-source is used, the effect can even be practically eliminated as shown in Fig. 9. A comparison of the integral ^{14}C-efficiencies with those obtained in a standard ^{14}C-window shows that, although the former may seem to offer better statistical accuracy, they are extremely sensitive to colour quenching, whereas the efficiency curves obtained for the window are practically insensitive to colour over a fairly wide range. Normally this range will be even more extended, as practical samples will hardly ever show so much variation in quenching properties as the two quenching agents used to obtain the curves shown.

It is an unfortunate aspect of liquid scintillation counting that optimalisation of measuring conditions has to be based, to a large extent, on trial and error methods. We feel that it would be worthwhile to refine the mathematical model sufficiently to be able to obtain more or less quantitative results from it. It could then become a useful tool for improving liquid scintillation methods and instrumentation. At that stage it would almost certainly become rewarding to study external standardisation by means of the model.

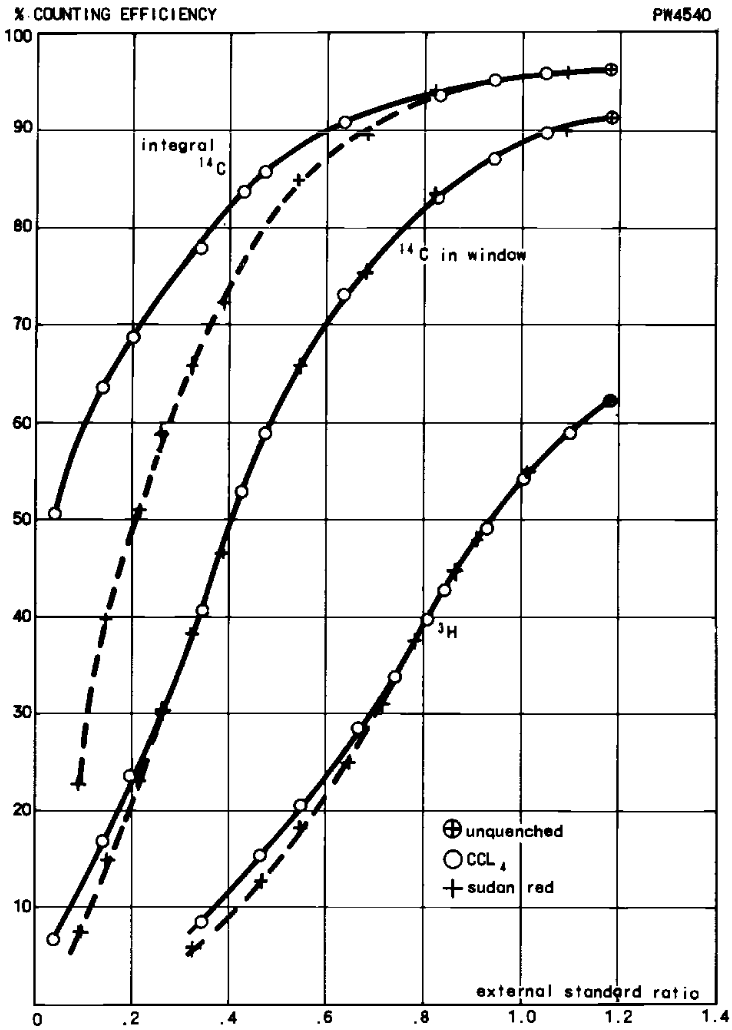


Fig. 9.

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3. P. E. Stanley and P. J. Malcolm, 'Practical liquid scintillation spectrometry: organising a methodology', this volume, pp. 44–62.
4. F. E. L. ten Haaf, in *Liquid Scintillation Counting,* Vol. 2 (eds. M. A. Crook, P. Johnson and B. Scales), Heyden, London, 1972, pp. 39-48.

DISCUSSION

L. A. Currie: By applying a log-transform to the abscissa (pulse height axis) you were able to compare relative pulse heights by simple translation (x-axis). Have you also applied such a transform to the ordinate (y-axis, pulse height frequency) in order to similarly compare the relative shapes of the pulse height frequency with distributions for unquenched, chemically quenched and colour-quenched samples?

F. E. L. ten Haaf: We did not do this, but it sounds a good idea. We might try it in the future.