

LIQUID SCINTILLATION COUNTING OF NOVEL RADIONUCLIDES

J.A.B. Gibson

Environmental & Medical Sciences Division,
AERE, Harwell, Oxon, UK

ABSTRACT

The theoretical background of counting radionuclides in liquid scintillators is presented. The effects of quenching and finite scintillator size are briefly described and the theory is justified by an experimental comparison between ^{55}Fe and ^3H in which all facets of the theory are important. Counting efficiencies for other nuclides decaying by 100% electron capture are calculated and compared with efficiencies for the β emitters ^3H , ^{14}C and ^{36}Cl . Also included are comments on the special problems associated with counting plutonium in biological materials. The essential conclusion is that in order to improve the technique and avoid unnecessary pitfalls it is necessary to have a sound understanding of the underlying theory of liquid scintillation counting.

INTRODUCTION

Liquid scintillation counting is a well established method for the measurement of a very wide range of radioactive isotopes. When a new radionuclide is used in an investigation the question arises as to whether it can be measured in a liquid scintillator. At this stage a series of questions should be asked.

- (i) What is the nature of its activity and is the decay scheme known so that it can be standardised and counting conditions can be optimised.
- (ii) Is there enough energy available for decay events in the scintillator to be detected by the photomultiplier.
- (iii) Is there a suitable compound of the isotope which can be incorporated into one of the many scintillator cocktails available, without undue quenching of the light output.

It is the answers to the first two questions that can be answered from an understanding of the basic interactions in the scintillator. The choice of scintillator cocktail is available in a wide range of reviews and conference proceedings eg (1-8).

Over the years the counting efficiency of instruments has increased and the background has been reduced as can be seen in table I(1). The counting efficiency for other isotopes emitting β^- or β^+ radiation has similarly improved and the efficiency for α emitting isotopes can be 100% in most cases. This leaves the important range of nuclides which decay by electron-capture (EC). In this decay it is X-rays and Auger electrons which must be measured in the scintillation counter and it is the measurement of these nuclides which will form the main basis of this paper.

TABLE I
Improvements in Instrument Performance(1)

	^3H		^{14}C	
	Counting Efficiency %	Background cpm	Counting Efficiency %	Background* cpm
1954	10	80	75	60
1962	25	55	80	30
1964	40	30	85	25
1969	60	20	90	16
1972	65	18	97	11

*Above ^3H end point.

In the paper I will present a brief introduction to the underlying theory of liquid scintillation counting, showing how the counting efficiency can be calculated for unquenched and quenched solutions of any isotope and how the theory is applied to EC nuclides. Comparisons with the counting efficiencies for β emitting isotopes will be made and the special problems of plutonium counting in biological material will be mentioned.

THEORY OF THE SCINTILLATION COUNTER

The theoretical approach in this section is a summary of a series of papers (9-13) and is specifically for single electron events in the scintillator as opposed to the theory for a β spectrum. The section includes a discussion of losses in the scintillator and photomultiplier, calculation of the counting efficiency, allowance for escape of X-rays from the scintillator without interaction, and the effects of quenching.

The essentials of the scintillation counter

The conversion of the initial electron energy into light photons is a complex process as detailed by Birks (14) and the essential details are set out in table II. The first important point is that the scintillation efficiency at high electron energies is about 4% for anthracene and for liquid scintillators may be about 1% as shown in table II.

TABLE II
Energy Transfer Processes in a Liquid Scintillator

Process	Energy Transfer	Gain 5 keV	Quanta keV ⁻¹		Quanta 5 keV
			1 MeV	5 keV	
Electron input	Energy deposition*		340	340	1730
Solvent Excitation	dE/dx losses at 5 keV	0.63	340	220	1080
Solute Excitation	Quenching and light losses				
Light Emission		0.01	3.4	2.2	10.8

*Assuming all the light is produced at 425 nm

Secondly at low energies, as shown by Gibson and Gale (10), following Birks (14), as the rate of energy deposition of a particle (dE/dx) increases then the light output per unit energy input is decreased i.e. the scintillation efficiency is reduced,

$$\frac{dL}{dE} = \frac{S(dE/dx)}{1 + kB(dE/dx)} \quad (1)$$

Where S is the scintillation efficiency and kB is a constant. The extent of this effect can be seen in figure 1 (10) in which the relative scintillation efficiency is plotted against electron energy.

The counting efficiency

The output from the scintillator is subject to normal statistical variations and at 1 MeV the distribution is conveniently approximated by a Gaussian distribution. At 5 keV with only 10.8 photons (on average) the distribution is a Poisson of the form,

$$G(r) = \frac{m^r e^{-m}}{r!}$$

where m is the mean number of photons and G(r) is the probability of having r photons in a given event. The 'zero probability' (9) is given when r = 0 i.e. G(0) = e^{-m}, and for m = 10.8, G(0) = 2.04 × 10⁻⁵. This is small and thus if we could count every photon then the counting efficiency for a 5 keV electron in the scintillator would be essentially 100%.

However, the efficiency of conversion of photons to electrons in a photomultiplier varies from 10 to 40%. If a is the conversion efficiency from photons to electrons

at the input to the first stage of the photomultiplier then the zero probability at this stage is (13)

$$Z = \exp\{-m(1 - e^{-a})\} \tag{2}$$

which approximates to $Z = e^{-am}$ when a is small. If we let $n = am$ then $Z = e^{-n}$ which is the zero probability for a Poisson distribution with a mean of n . The effect of later stages in the photomultiplier are small (<1%) if the first stage gain exceeds 5. The counting efficiency is $\epsilon = 1 - Z$. Using the data from table II the counting efficiency is given in table III. It should be noted that this is the maximum achievable efficiency at zero bias level on the discriminator following the amplifier and in practice lower efficiencies will be obtained.

The efficiency for two photomultiplier tubes in coincidence is slightly more complicated in that at least two photons must be produced and one photon must be detected by each photomultiplier. The distribution is of the form

$$G(r) = (1 - 0.5^{r-1}) n^r \frac{e^{-n}}{r!} \tag{3}$$

where $n = m(1 - e^{-a})$ as before. Equation (3) only applies where $r \geq 2$ and the counting efficiency for the two tubes in coincidence, ϵ_c is simply the product of the counting efficiencies of the single tubes (ϵ_s). If the two tubes have equal counting efficiencies then $\epsilon_c = \epsilon_s^2$ as given in table III. In order to determine the counting efficiency for a β spectrum it is necessary to integrate over the whole spectrum (13).

The use of the coincidence system greatly reduces the background but it also reduces the counting efficiency and these two effects have to be balanced in a given application.

TABLE III
Counting Efficiency for Single and Coincidence
Systems for 5 keV Electrons

Photocathode Efficiency, a %	Mean no. of electrons $n = am$	Zero Probability		Efficiency %	
		e^{-n}	Equ ⁿ (2)	$\epsilon_s = 1 - Z$ Single	ϵ_s^2 Coincidence
10	1.08	0.340	0.358	64.2	41.2
20	2.16	0.115	0.141	85.9	73.8
30	3.24	0.039	0.061	93.9	88.2
40	4.32	0.013	0.028	97.2	94.4

Figure of merit

It is useful to specify an overall efficiency for the conversion of input of electron energy into a mean numbers of electrons at the first dynode of the photomultiplier $n(E)$. This is defined as

$$P = \frac{n(E)}{E \cdot F(E)} \quad (4)$$

where $F(E)$ is the relative scintillation efficiency as given in figure 1. P is independent of energy E and for the system described in tables II and III, $P = 3.4 \times 10^{-3} \text{ e keV}^{-1}$ at a photocathode efficiency of 10%. It is possible to measure P by various methods as discussed by Gibson (13).

The effects of quenching

A major problem in liquid scintillation counting is a reduction in the scintillation efficiency due to impurities introduced with the sample. This quenching effect can take two main forms (a) chemical and (b) colour quenching.

Chemical quenching involves the interception of energy in the transfer and diffusion processes between molecules. It can be thought of in simple terms as follows

- (i) Energy absorption $M + E \rightarrow M^*$ Rate = 1
- (ii) Light emission $M^* \rightarrow M + h\nu$ Rate = $k_1 [M^*]$
- (iii) Quenching $M^* + Q \rightarrow \text{Loss}$ Rate = $k_2 [M^*][Q]$

where M is a fluorescent molecule in the ground state
 M^* is a fluorescent molecule in an excited state
 Q is a quenching molecule
 $[]$ are molecular concentrations
 $k_{1,2}$ are relative reaction rate constants.

Then the total available fluorescence is

$$S_0 = k_1 [M^*] + k_2 [M^*][Q]$$

and fluorescence from a quenched solution is

$$S = k_1 [M^*]$$

The relative quenching factor is then

$$g = \frac{S}{S_0} = \frac{1}{1 + kw} \quad (5)$$

where $k = k_1/k_2$ and $w = [Q]$. Equation (5) is a Stern-Volmer equation and is very similar to the equation (1) for ionisation quenching in the track of the ionising particles where kw is replaced by $kB(dE/dx)$. The factor g for chemical quenching is independent of electron energy and reduces the figure of merit from P to gP and thus

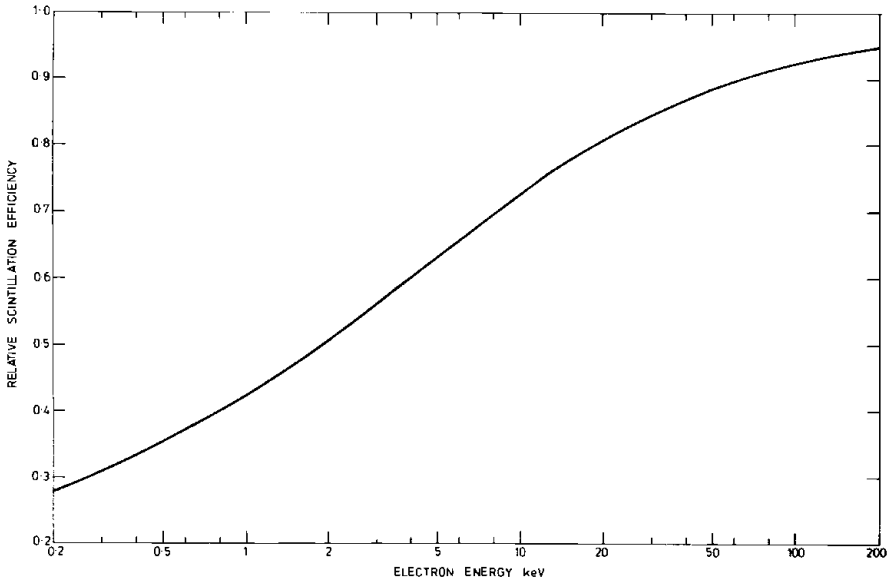


FIG.1.EFFECT OF IONISATION QUENCHING ON THE SCINTILLATION EFFICIENCY

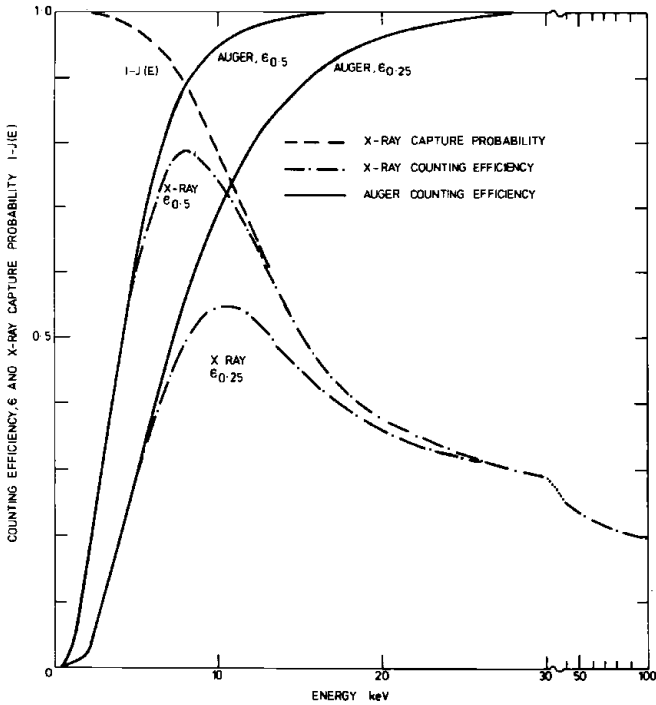


FIG.2.CALCULATED COUNTING EFFICIENCY FOR AUGER ELECTRONS AND X-RAYS

changes $\exp(-n)$ to $\exp(-gn)$ and so reduces the counting efficiency. This is discussed more fully elsewhere (11).

Colour quenching involves an interception of the light-photons and may follow equation (5). At high quench levels it will distort the spectrum and there is no really satisfactory theory to predict the changes in counting efficiency, eg ten Haaf (15).

Loss of energy into the scintillator walls

The discussion has so far assumed that the scintillator is infinite in volume and for electrons this is a reasonable assumption for volumes of about 15 cm^3 at energies up to 100 keV. Even above this energy enough energy is deposited in the scintillator to produce a pulse in the photomultiplier and so the counting efficiency would be 100%. However for X (and γ) rays there is a high probability of escape from the scintillator without any interaction at all. This escape probability is given by $e^{-\mu d}$ where d is the distance the X-ray travels in the scintillator. Integrating $e^{-\mu d}$ over all solid angles for a cylindrical volume the total probability of escape $J(E)$ can be calculated and the capture probability $(1 - J(E))$ determined (figure 2 (16)). This capture probability is multiplied by the counting efficiency for electrons to obtain a counting efficiency for X-rays in a given scintillator (figure 2).

COUNTING OF NUCLIDES DECAYING BY ELECTRON CAPTURE

Electron capture is a mode of nuclear decay in which K or L electrons are captured by the nucleus resulting in a vacancy in one of these shells. In the subsequent rearrangement of the electrons within the shells either fluorescent X-rays or Auger electrons will be emitted depending upon the atomic number of the daughter nuclide. The X-ray can escape from the scintillator and if the Auger electrons have very low energies then dE/dx effects are very important in calculating the counting efficiency. The number of possible interactions is very large and ignoring capture in the M and higher shells and assuming a single energy for all shells then 15 interactions are possible as shown in figure 3. However many of the interaction probabilities are small and taking ^{55}Fe as an example the 5 most important are given in table IV. In the table the coincidence counting efficiency, ϵ_i , for each interaction is

$$\epsilon_i = \phi_i (1 - e^{-n})^2$$

where ϕ_i is the interaction probability. More details are given elsewhere (16). If one assumes that all the energy is deposited at 5.89 keV ignores the dE/dx effect then $\epsilon_c = 0.898$ and allowing for the dE/dx effect $\epsilon_c = 0.734$ both of which are significantly greater than the value in the table of $\epsilon_c = 0.644$. The theoretical result will be compared with experimental data below.

TABLE IV
Calculation of the Coincidence Counting Efficiency
for ^{55}Fe with $P = 0.5 \text{ e keV}^{-1}$

Interaction	Electron Energy keV	dE/dx Factor F(E)	E.F(E).P n	ϵ'_i	ϕ_i	ϵ_i
(i) K X escape						
L Auger	0.64	0.378	<u>0.121</u>	0.013	0.013	0.000
(ii) K X capture	5.61	0.650	1.823			
L Auger	0.64	0.378	0.121			
C Auger	0.28	0.307	<u>0.043</u>			
			<u>1.987</u>	0.745	0.267	0.199
(iii) K Auger	5.24	0.641	1.679			
L Auger	0.64	0.378	0.121			
L X capture	0.37	0.330	0.061			
C Auger	0.28	0.307	<u>0.043</u>			
			<u>1.904</u>	0.724	0.001	0.001
(iv) K Auger	5.24	0.641	1.679			
L Auger	0.64	0.378	0.121			
L Auger	0.64	0.378	<u>0.121</u>			
			<u>1.921</u>	0.729	0.608	0.443
(v) L Auger	0.67	0.382	0.128	0.014	0.110	0.001
					0.999	0.644

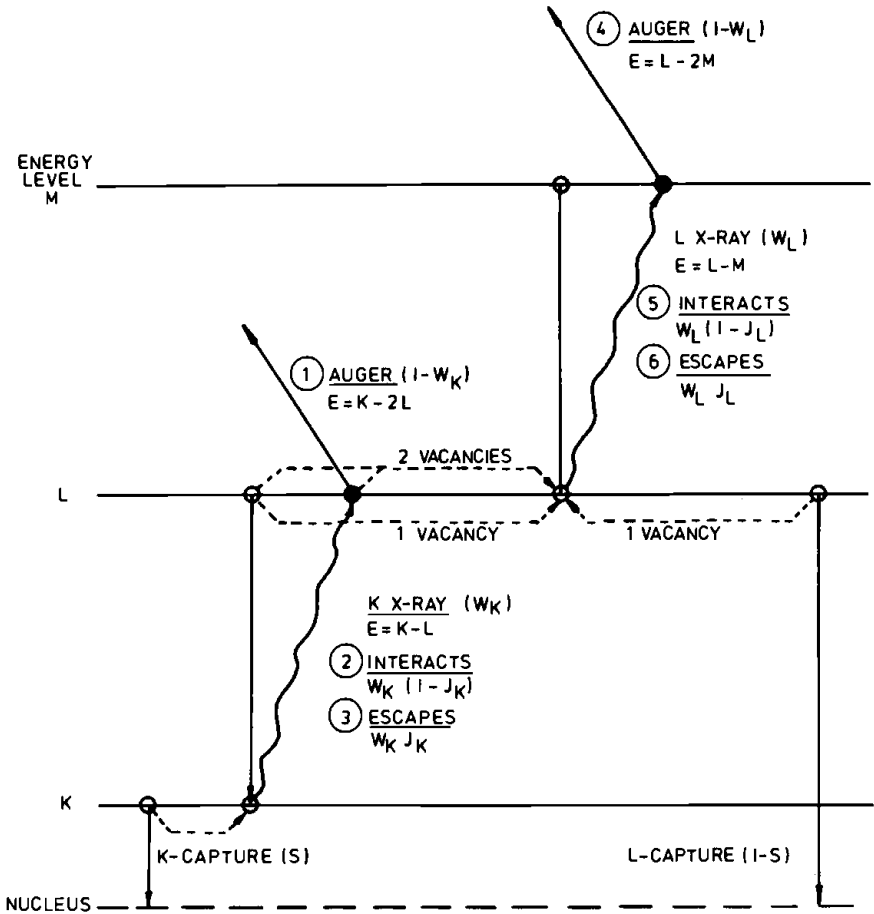
$$\text{Efficiency, } \epsilon_i = \phi_i \epsilon'_i$$

Experimental comparison between ^{55}Fe and ^3H

In order to establish the theory discussed above it was decided to examine the apparent paradox that the counting efficiency for ^{55}Fe (ϵ_{Fe}) can be greater or less than that for ^3H (ϵ_{H}) for different quenching conditions (or values of P). The theoretical technique is as discussed above for ^{55}Fe and as discussed elsewhere (10) for ^3H . The experimental technique was as follows. Two alternative scintillator solutions were used (a) 14 cm³ of NE220 (Nuclear Enterprises) + 0.1 cm³ of standard solution and (b) 14 cm³ of BBS-toluene mixture + 1 cm³ of standard solution. The counting efficiency for $^3\text{H}_2\text{O}$ was 35% and 28% respectively for the two scintillators. $^{55}\text{Fe}(\text{NO}_3)_3$ was dissolved in 0.1N HNO₃. The standard solutions were obtained from TRCL (Amersham) and have standard errors on the mean of 1.4% for ^{55}Fe and 1.3% for ^3H . The counting efficiency of the solutions was reduced by adding successive small aliquots of acetone.

All experimental measurements were made with a Packard Tri-Carb (Model No. 3003) at a temperature of 2°C. Integral bias curves were obtained and extrapolated to zero

COUNTING NOVEL RADIONUCLIDES



		NO OF INTERACTION
K CAPTURE	① AUGER + 2L VACANCIES	6
	② K-X-RAY* + 1L VACANCY	3
	③ ESCAPE + 1L VACANCY	3
L CAPTURE†	④ AUGER + 2 M ELECTRONS	1
	⑤ L X RAY + 1M + 1C ELECTRON	1
	⑥ ESCAPE + 1M ELECTRON	1
	TOTAL	15

* PHOTOELECTRON OF ENERGY $K - L - C$

C = CARBON ELECTRONS

† AND L VACANCIES FROM ①, ② AND ③

W = X RAY FLUORESCENT YIELD

FIG.3. SCHEMATIC DIAGRAM OF ASSUMED INTERACTIONS IN ELECTRON CAPTURE

bias. An automatic external standard was used to check that the counting efficiency for ^{55}Fe and ^3H was the same after the addition of the acetone aliquot and if necessary small corrections ($< 2\%$) were made.

The ratio of the counting efficiencies ($\epsilon_{\text{F}}/\epsilon_{\text{H}}$) are plotted in figure 4 as a function of ϵ_{H} . The ratio is for either the NE220 solution using both isotopes in separate vials or for the BBS-toluene mixture again using both isotopes. Only the calculated counting and volumetric standard deviation is shown on the figure and there is an additional systematic standard error on the mean of $\pm 2\%$ due to uncertainties in the standards. The solid line in figure 4 is that obtained from the theoretical calculations discussed above. The line has not been normalised. The agreement between the theoretical and experimental results in figure 4 is very good and supports the approach to calculating counting efficiencies proposed by the author. It is seen from the curve that the effect of quenching on the $\epsilon_{\text{F}}/\epsilon_{\text{H}}$ ratio depends upon the figure of merit of the system and it is only for a single tube with $P > 0.55\text{e keV}^{-1}$ that $\epsilon_{\text{F}}/\epsilon_{\text{H}}$ will increase as quenching is increased.

The reason for the change in slope of the curve is that at high counting efficiencies all the electrons produced by ^{55}Fe (figure 2) will be counted and if, for example, $n = 6$ then $\epsilon_{\text{F}}' = (1 - e^{-6})^2 = 0.995$. With a large chemical quenching factor, (equation (5)) eg $g = 0.5$ then $n = 0.5 \times 6$ and $\epsilon_{\text{F}}' = 0.902$ which is only a small change (9%) in efficiency. (The counting efficiency will be reduced due to the escape of K.X-rays and the above figures for ϵ_{F}' should be multiplied by 0.875 to obtain the true counting efficiency, ϵ_{F}). However for ^3H , about half of the β rays have energies less than 5.9 keV and these will be more susceptible to changes by quenching resulting in a more rapid reduction in counting efficiencies ($\epsilon_{\text{H}} < 0.6$) than the counting efficiency for $n = 2$ is $\epsilon_{\text{F}}' = 0.75$ and for $g.n = 1$, $\epsilon_{\text{F}}' = 0.40$ (again both are to be multiplied by 0.875). In this case the β particles below 5.9 keV for ^3H are only a minor contributor to ϵ_{H} and so ϵ_{H} reduces more slowly than ϵ_{F} for increasing levels of quenching. Thus depending on the figure of merit for the system the ratio $\epsilon_{\text{F}}/\epsilon_{\text{H}}$ can increase or decrease with the changes in quenching levels.

Horrocks (17) suggested that for the BBS-toluene mixture the ^{55}Fe may not be dissolved in the scintillator but it is dispersed as sols. These droplets would discriminate against the low energy Auger electrons and so reduce the ^{55}Fe counting efficiency relative to that for the higher energy β particles of ^3H . Experiments to measure the droplet size showed that at the concentrations used (0.6% W/W) they were less than 10 nm in diameter. Advice from sol physicists and information in the literature (18, 19) suggests that the droplet size will not be greater than 5 nm. The range of electrons in water is as given below.

Electron Energy keV	0.28	0.65	5.24	5.9
Electron Range nm	7	28	830	1000

COUNTING NOVEL RADIONUCLIDES

Thus it is suggested that only the lowest energy electrons will be effected and the similarity between the separate results for the two scintillators and with the theory tends to confirm this hypothesis.

COUNTING OF OTHER RADIONUCLIDES

There are a range of EC nuclides and Horrocks (20) suggested a list which could be useful in tracer studies etc. This list is given in table V together with their calculated counting efficiencies. The reduction in counting efficiency for ^{97}Tc is due mainly to the escape of K X-rays as the fluorescent yield is 73% for Mo. Above this atomic number the fluorescent yield increases to 97% for Tl but the L X-rays (and Augers) are counted with increasing efficiency. Corresponding counting efficiencies for ^3H , ^{14}C and ^{36}Cl are included for comparison and efficiencies for any other isotope with a known decay scheme (and β spectrum) can be calculated by this method.

TABLE V
Counting Efficiencies for EC and β^- Nuclides
for $P = 0.50$ and 0.25e keV^{-1}

Nuclide	Half-life	K_{α} X-ray or β max Energy keV	K capture Probability	Coincidence Counting Efficiency %	
				$P = 0.5$	$P = 0.25$
^{37}Ar (EC)	35d	2.6	0.91	23	8
^{55}Fe (EC)	2.6a	5.9	0.89	64	34
^{71}Ge (EC)	11.4d	9.2	0.88	76	55
^{97}Tc (EC)	$2.6 \times 10^6\text{a}$	17.5	0.84	57	47
^{131}Cs (EC)	9.7d	29.8	0.87	70	52
^{179}Ta (EC)	600d	55.8	0.63	87	53
^{205}Pb (EC)	$3 \times 10^7\text{a}$	12.2	*	95	72
^3H (β)	12.26a	18.6	-	57	35
^{14}C (β)	5730a	156.	-	95	91
$^{36}\text{Cl}^+$ (β)	$3.1 \times 10^5\text{a}$	714.	-	99	98

*Only 35 keV available for the decay.

+1.9% EC $K_{\alpha 1} = 2.3\text{ keV}$.

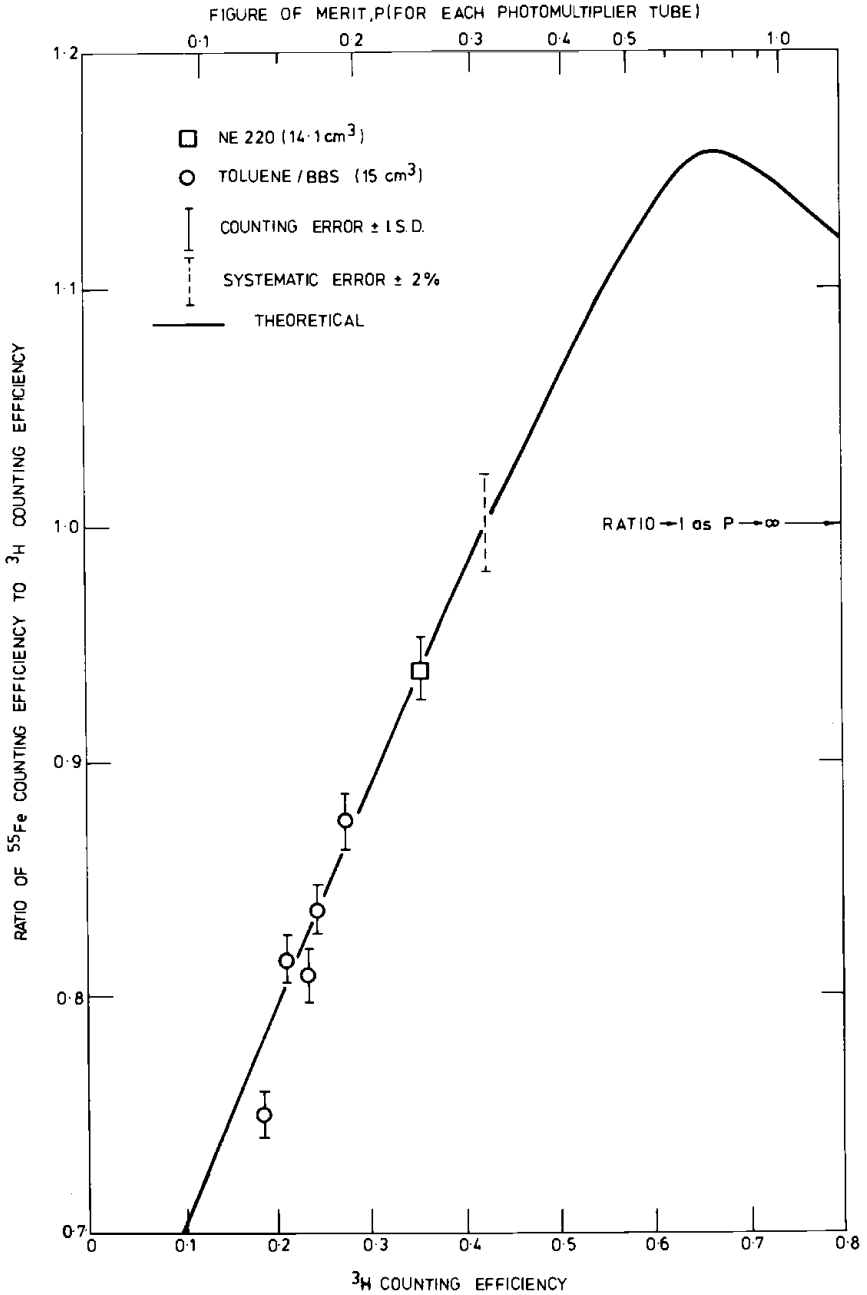


FIG.4. RATIO OF ⁵⁵Fe/³H COUNTING EFFICIENCIES FOR A COINCIDENCE SYSTEM

Determination of plutonium alpha activity and ^{241}Pu in biological materials

This is a particular problem of the nuclear energy industry where levels of soluble plutonium in man can only be determined by urine and faecal counting. The additional problem is the increasing levels of ^{241}Pu due to increased burn-up of fuel in reactors. ^{241}Pu is a β -emitter ($E_{\text{max}} = 20.8 \text{ keV}$, $T_{1/2} = 13.2\text{a}$). After extraction of the plutonium from the biological material then liquid scintillation counting offers a convenient method of determining both the α activity and the ^{241}Pu β activity simultaneously.

Horrocks and Studier (21) first described the technique for ^{241}Pu in urine. Eakins and Lally (22) have a relatively simple technique of gel scintillation counting for Pu α and β^- activity determination. The α activity from ^{239}Pu (+ ^{238}Pu and ^{240}Pu) can be counted with 100% efficiency but it is convenient to reduce it to 90% to count the ^{241}Pu at the same time with a counting efficiency of 21%. This technique is used to detect 1 pCi of α activity and 10 pCi of ^{241}Pu at the background level. The limits of detection are about 20% of these levels. The technique is adequate for the ^{241}Pu counting in urine, faeces and nose blow samples but to obtain adequate sensitivity for ^{239}Pu in urine (0.02 pCi) it is still necessary to measure electrodeposited sources in a solid-state counter.

CONCLUSION

Liquid scintillation counting is the accepted technique for many radio isotopes in a wide range of chemical forms. It lacks sensitivity for very low levels of α -activity and cannot compete directly with the internal gas counter for natural levels of tritium. The technique has come a long way since the first major conference recorded by Bell and Hayes in 1958 (7). It is as important now as it was 20 years ago to establish an understanding of the underlying theory behind the techniques so that improvements can be made and pitfalls avoided. Electron capture nuclides are valuable tools in all types of tracer techniques but they can produce unusual results unless a proper understanding is obtained.

REFERENCES

1. D.L. Horrocks, Applications of Liquid Scintillation Counting. New York and London: Academic Press (1974).
2. A. Dyer, Ed, Liquid Scintillation Counting, Vol 1. London, New York and Rheine: Heyden & Son Ltd. (1971).
3. M. Crook, P. Johnson and B. Scales, Ed, Liquid Scintillation Counting, Vol 2. London, New York and Rheine: Heyden & Son Ltd. (1972).
4. M.A. Crook and P. Johnson, Ed, Liquid Scintillation Counting, Vol 3. London, New York and Rheine: Heyden & Son Ltd. (1974).
5. E.D. Bransome, Ed, The Current Status of Liquid Scintillation Counting, New York and London: Grune and Stratton (1970).

6. D.L. Horrocks and C-Z. Peng, Ed, *Organic Scintillators and Liquid Scintillators*. New York and London: Academic Press (1971).
7. C.G. Bell and F.N. Hayes, Ed, *Liquid Scintillator Counting*. London, New York, Paris and Los Angeles: Pergamon Press (1958).
8. J.A.B. Gibson and A.E. Lally, *Analyst* 96, 681 (1971).
9. H.J. Gale and J.A.B. Gibson, *J. Sci. Instrum.* 43, 224, (1966) and AERE Report R 507 (1965).
10. J.A.B. Gibson and H.J. Gale, *J. Sci. Instrum. (J. Phys. E) Ser 2*, 1, 99 (1968).
11. J.A.B. Gibson and H.J. Gale, *Int. J. Appl. Rad. & Isotopes*, 18, 681, (1967).
12. J.A.B. Gibson *in* *Liquid Scintillation Counting*, Vol 2, p 23. (M.A. Crook, P. Johnson and B. Scales, Ed). London, New York and Rheine: Heyden and Sons (1972).
13. J.A.B. Gibson. AERE Report R 6919 (1972).
14. J.B. Birks. *The Theory and Practice of Scintillation Counting*, Oxford. Pergamon Press (1964).
15. F.E.L. ten Haaf *in* *Liquid Scintillation Counting*, Vol 2, p 39, (M.A. Crook, P. Johnson and B. Scales, Ed). London, New York and Rheine: Heyden & Son (1972).
16. J.A.B. Gibson and M. Marshall. *Int. J. Appl. Radiat. & Isotopes*, 23, 321, (1972).
17. D.L. Horrocks, Private communication.
18. S. Glasstone, *Text book on Physical Chemistry* p 1267. London: MacMillan, (1948).
19. N.K. Adam, *Physical Chemistry*, p 609. Oxford: Clarendon Press (1958).
20. D.L. Horrocks. *Int. J. Appl. Radiat. & Isotopes*, 22, 258, (1971).
21. D.L. Horrocks and M.H. Studier, *Anal. Chem.* 30, 1747, (1958).
22. J.D. Eakins and A.E. Lally *in* *Liquid Scintillation Counting*, Vol 2, p 155 (M.A. Crook, P. Johnson and B. Scales, Ed). London, New York and Rheine: Heyden & Son (1972).