

Chapter 2

The Absolute Method of Measurement of Carbon-14 Activity by 4π Liquid Scintillation Counter

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INTRODUCTION

Standardisation of radionuclides by the use of the 4π liquid scintillation counter is possible due to the full geometry of counting and due to the absence of the main counting corrections such as self-absorption in the source and absorption in the source holder. In the case of carbon-14, as a low energy beta emitter, the so called 'cut off energy' of the counter should be taken into account. It determines the lowest energy of the beta particle which is needed to be counted by the scintillation counter. Here the results of measurements of n-hexadecane labelled with carbon-14 are given, which was counted in single channels, in the coincidence and in the parallel systems.

THE COUNTING EQUIPMENT

All measurements have been made in the home-made scintillation head shown in Fig. 1. It contains the glass vial (12 ml in volume), a light pipe made of perspex and two photomultipliers (EMI 9514S).

The electronic equipment shown in Fig. 2 enabled counts to be made in the single channels, in the coincidence, and in the parallel system (as a sum) with variable levels of discrimination (from 35 to 145 mV, before amplification). It contains two cathode followers, two discriminators, two amplifiers (with a fixed gain of amplification 275 V/V) the coincidence unit (type A 491-A 422 Rochar Electronique) with a resolving time of $1\mu\text{s}$, the sum unit, the quenching unit with a regulated dead time from 5 to $150\mu\text{s}$ and the scaler.

PPO in toluene (3 g/l) and PBD in toluene (10 g/l) have been used as scintillators. The normal volume of scintillator used in all counting was 5 ml. All measurements were carried out with the scintillation head placed in a refrigerator at a temperature of -20°C .

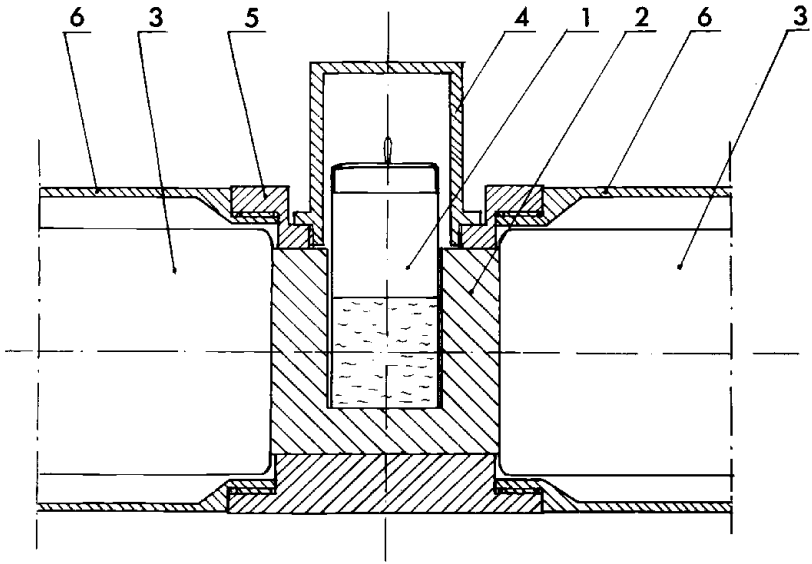


Fig. 1: The double scintillation head. 1. Counting vials with scintillator and a sample. 2. Light pipe made of perspex. 3. Photomultipliers. 4. Sleeve covering the counting vial. 5, 6. Body of the scintillation head, shielding the light pipe and photomultipliers.

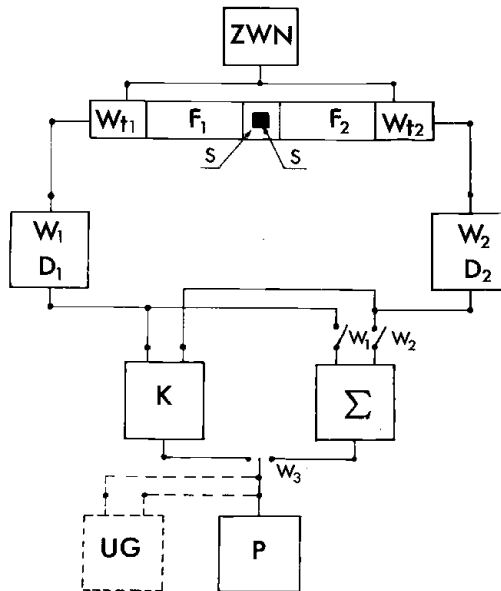


Fig. 2: Block diagram of the counting equipment. ZWN: E.h.t. unit. F: Photomultipliers. W_t : Cathode followers. W, D: Discriminator with amplifier. K: Coincidence unit. Σ : Sum unit. P: Scaler. U.G.: Quenching unit.

THE COUNTING METHOD

Several counting vials with 5 ml of scintillator in each and a small amount of n-hexadecane labelled with carbon-14 (supplied by The Radiochemical Centre, Amersham) were prepared. The quantity of the radioactive material in each sample was determined by weighing. First, the anode characteristics were checked to choose the proper e.h.t. value as the working parameter.¹ The anode characteristics of the separate channel showed some displacement owing to the different sensitivities of the photomultipliers. In spite of this the anode characteristics for the parallel system and for coincidence were taken for equal e.h.t. value in the channels. The characteristics shown in Fig. 3 were taken for a discrimination level of 35 mV and for two different values of the dead time (for the single channel and the parallel system), i.e. 5 and 50 μ s.

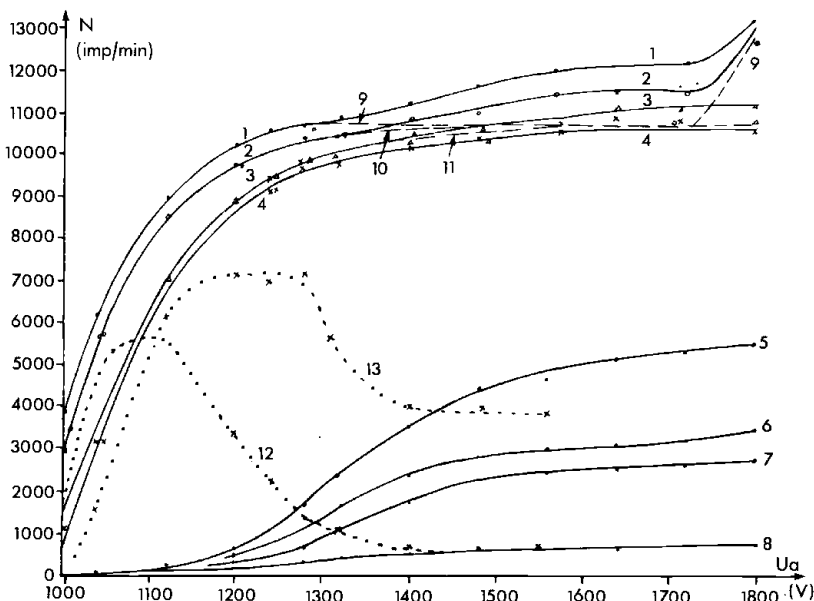


Fig. 3: The anode characteristics of the 4π liquid scintillation counter for 35 mV of discrimination level. 1, 2, 3, 4: Characteristics (after subtracting background) for the parallel system, channel I, channel II and the coincidence system. 5, 6, 7, 8: Background characteristics for the parallel system, channel I, channel II and for the coincidence system. 9, 10, 11: Characteristics for the parallel system, channel I and channel II with the quenching unit of 50 μ s dead time. 12, 13: The counting rate of a sample to background ratio $\frac{N}{N_C}$ for the parallel and coincidence system.

To show the influence of e.h.t. value on the results of counting, the discrimination characteristics were taken for coincidence and parallel systems for two different values of e.h.t. and are shown in Fig. 4. The value of 1320 V was chosen and fixed for further counting because it ensured the highest possible counting efficiency.

The counting rate of the background from the scintillation was, of course, higher in the parallel system than in the remaining systems and it was kept at a level of 2000 counts/min by the low temperature of the refrigerator (-20°C).

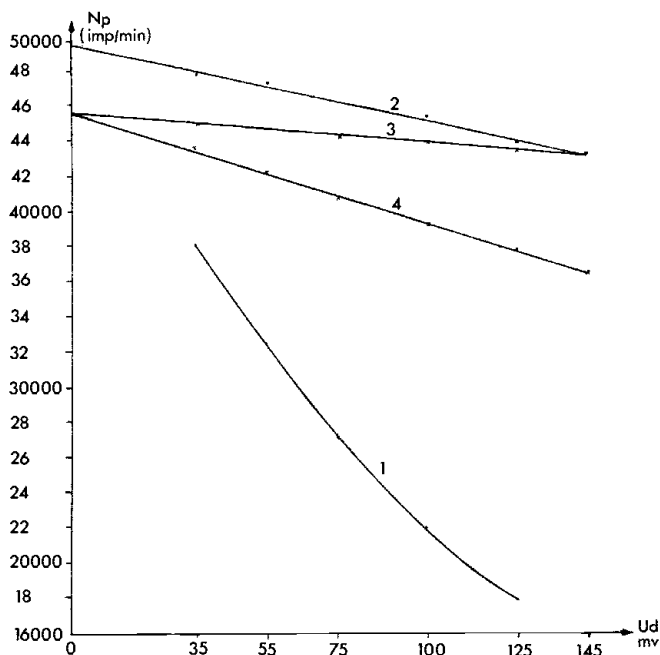


Fig. 4: The discrimination characteristics of the 4π counter. 1, 2: Curves for the parallel system for anode voltage 1120 V and 1320 V correspondingly. 3, 4: Curves for the coincidence system for anode voltage 1320 V and 1560 V correspondingly.

All sources were counted with a variable discrimination level (from 35 to 145 mV) to obtain the discrimination curves and to extrapolate the counting rate to the zero level of discrimination.

No counting corrections were applied for the calculation of the radioactive concentration of the counting n-hexadecane except for the correction due to the background of the counter and for the dead time.

RESULTS OF COUNTING

To measure the radioactive concentration of n-hexadecane, 7 samples were prepared and counted in the manner described previously, using the parallel system. The results are shown in Table 1.

Table 1. The results of radioactive concentration for n-hexadecane labelled with carbon-14, counted in the parallel system

No. of sample	Mass of sample (g)	Scintillator	Radioactive Concentration ($\mu\text{Ci/g}$)	
1	0.02203	PBD	1.075	Mean value
2	0.02155	PBD	1.079	
3	0.02178	PPO	1.062	1.072 $\mu\text{Ci/g}$
4	0.02133	PBD	1.091	
5	0.02106	PPO	1.060	standard error
6	0.02144	PBD	1.083	
7	0.01712	PPO	1.058	

The obtained mean value for the radioactive concentration was

$$1.072 \pm 0.005 \mu\text{Ci/g}$$

The figure given by the Radiochemical Centre was

$$1.07 \mu\text{Ci/g}$$

Apart from counting in parallel systems, 4 samples were prepared and counted in all three systems to compare the counting efficiency. The results are given in Table 2.

Table 2. Comparison of the counting efficiency for the parallel, coincidence and the single channel systems for n-hexadecane labelled with carbon-14 (the results in the parallel system are taken as 100%).
Scintillator—PBD in toluene.

No. of sample	Parallel system $\mu\text{Ci/g}$	Coincidence system efficiency %	Channel I efficiency %	Channel II efficiency %
1	1.059 mean value	93.6 mean value	95.6 mean value	96.2 mean value
2	1.087	93.4	97.0	97.9
3	1.084 <u>1.073</u> $\mu\text{Ci/g}$	92.8 <u>93.4</u>	96.3 <u>96.2</u>	96.9 <u>97.0</u>
4	1.061	93.7	95.9	98.8

Compared to the parallel system, the obtained mean value of the counting efficiency was 93.4% for the coincidence system and 96.2%; 97.0% for the two single channels.

THE SECONDARY PULSES

The anode characteristics from Fig. 3 showed some discrepancy in the case of two different dead time values ($5\mu\text{s}$ and $50\mu\text{s}$). This discrepancy can be explained by secondary pulses arising at the scintillation counter after the main pulses. It should be pointed out that the secondary pulses are observed for the single channel and the parallel system counting but not for a coincidence. The time distribution function of the quantity of these pulses is demonstrated in Fig. 5. This time distribution function has been measured by the electronic equipment (shown in Fig. 2) using a coincidence unit with the addition of a delay unit in one channel. The principle of measurement depended upon coincidence cases between the main (true) pulse in one channel and the secondary pulses, arising after the main pulse, in the second channel, with a variable time of delay. As it is shown in Fig. 5, there is a constant counting rate of the 'true' coincidences as a function of the delay time from 0 to $0.8\mu\text{s}$. This is due to the resolving time of the coincidence unit, which amounts to $1.05\mu\text{s}$ and the minimum delay time of the delay unit which amounts to $0.25\mu\text{s}$. The counting rate rapidly decreased for a further delay and increased again

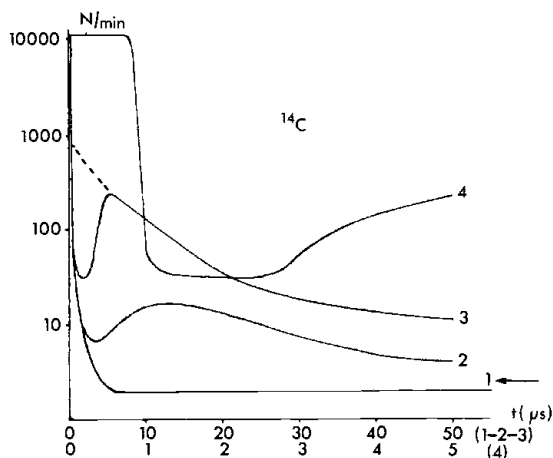


Fig. 5: Time distribution of secondary pulses. 1: Calculated level of the random coincidences between main ("true") pulses. 2: Time distribution for anode voltage 1400 V. 3, 4: Time distribution for anode voltage 2000 V for a delay of 50 μ s (3) and 5 μ s (4).

after exceeding 3 μ s of delay time. This is due to the dead time in the channels which amounts to about 3 μ s. Thus the optimum counting rate has been obtained which gives a rather false picture of the phenomena. One may conclude that the true time distribution should be extrapolated to the zero level of delay and in any case the period from 0 to 5 μ s cannot be properly investigated by this method. For a period from 5 to 50 μ s the counting rate is reduced, but even for 50 μ s it does not obtain the level of a random coincidence rate.

The random coincidence rate has been calculated from the equation:

$$N_R = 2 \tau_R N_1 N_2$$

where τ_R is the resolving time, N_1 , N_2 are the counting rates in the channel 1 and channel 2.

Taking this into account one can conclude that the curves 2, 3, 4 (Fig. 5) show the time distribution of secondary pulses. It should be emphasised that the amount of secondary pulses does depend upon the e.h.t. value. Further experiments which were carried out, for other radionuclides, lead to the conclusion that the amount of secondary pulses depends also on the energy of β -particles. For example for phosphorus-32 more secondary pulses were observed than for carbon-14.

The reasonable explanation of the origin of the secondary pulses, with time distribution longer than 50 μ s, is rather difficult. First of all, it should be observed that the secondary pulses arise for a total gain of amplification (photomultiplier + amplifier) high enough to count the single electron from the photocathode.

The second conclusion is that such a long period in time distribution excludes the origin of secondary pulses, as a result of imperfect vacuum in the photomultiplier tube, as the ions from the rest of the gases in the tube should arrive at the electron in a period comparable to 1 μ s. Providing that the electronic circuit does not cause the secondary

pulses (as proved for the circuit in question), the only explanation which the author can suggest is the phosphorescence effect of light from the transparent material such as perspex used as a light pipe, glass of the counting vial and especially the glass envelope of the photomultiplier tube.

CONCLUSIONS

The main conclusion which can be made from the experiments described above is that the 4 π liquid scintillation counter can be used as the absolute method for standardisation of carbon-14 compounds. Full efficiency of counting (100%) has been obtained in a limit of the counting errors. According to Horrocks and Studier,² the theoretical counting efficiency can be calculated providing that the minimum energy needed for producing one single electron from the photocathode is given. Taking carbon-14, they obtained 99.1% for single channel and 97.1% for coincidence, providing 1 keV as a minimum energy needed and 98.1% for single channel and 93.9% for coincidence providing 2 keV as a 'cut-off energy'. Taking the liquid scintillator efficiency as 70% to anthracene (with 4% of absolute efficiency) and 15% efficiency for photocathode, one may obtain about 620 eV as a minimum energy needed for one single electron, providing that all photons produced by the scintillator strike the photocathode (i.e. full light collection). Taking into account that full light efficiency is never obtained in the scintillation head, but the highest possible efficiency is obviously in the parallel system of counting, it is worthwhile to compare the theoretical data with the obtained results. Concerning the secondary pulses, simply demonstrated in the paper as a possible source of counting errors, one can overcome this problem by applying the quenching unit at high total gain of amplification, or simply by counting in the coincidence system.

REFERENCES

- 1 T. Radoszewski, *Nukleonika* 5, 361 (1960).
- 2 D. H. Horrocks and M. H. Studier, *Anal. Chem.* 33, 615 (1961).
- 3 J. B. Birks, *The Theory and Practice of Scintillation Counting*, Pergamon Press, London, 1964.
- 4 T. Radoszewski, *Nukleonika* 7, Part 7, to 8 (1962).

DISCUSSION

J. B. Birks: The most probable origin of the secondary pulses is the diffusion-controlled interaction of pairs of triplet-excited solute molecules, leading to the delayed scintillation component. This component is not readily observed in liquid solutions containing oxygen, but the use of low temperature solutions reduces the oxygen solubility and thus reduces oxygen quenching of the triplet states. Your results are similar to those of Voltz and Laustriat at Strasbourg.

T. Radoszewski: I agree with you but we have observed very similar effects when we illuminated just the photomultiplier with light pulses from the light source.

J. A. B. Gibson: The theoretical efficiency at zero bias is always less than 100% and quenching will reduce this efficiency still further. The correction is of the order of 1 or 2% for carbon-14 but for lower energy β -emitters this correction will increase to 30% or

more (for tritium, etc.). This correction may be obtained as outlined by J. A. B. Gibson and H. J. Gale, *J. Sci. Instrum. (Physics E)*, Ser. 2, 1, 99 (1968) (See Fig. 6) and V. Kolarov, Y. Le Gallic and R. Vatin, *Intern. J. Appl. Radiation Isotopes* 21, 443 (1970).

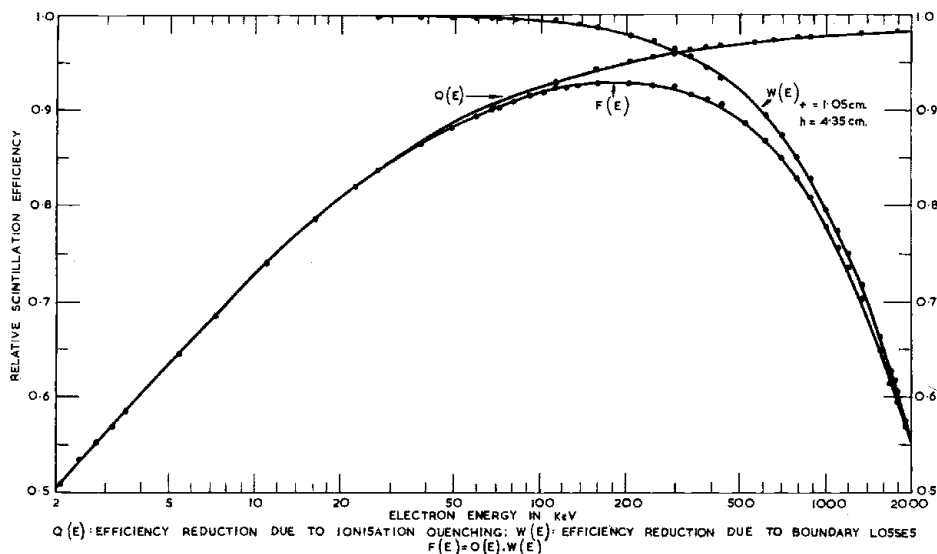


Fig. 6: Variation of scintillation efficiency with electron energy. [From J. A. B. Gibson and H. J. Gale, *J. Sci. Instrumen. (Physics E)* Ser. 2, 1, 99 (1968)]

T. Radoszewski: As I said, the theoretical efficiency for carbon-14 has been calculated and it amounts to about 99%; therefore the practical full efficiency of counting could be obtained in a limit of error.

D. A. Kalbhen: Are secondary pulses different with other scintillator solutes than PPO and POPOP, or other glass vials or other optical coupling materials?

T. Radoszewski: I did not find any difference between PPO and PBD in toluene.