

Chapter 6

The 4π Liquid Scintillation Method for Activity Measurement of Electron Capture Nuclides

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INTRODUCTION

The activity of electron-capture nuclides have been previously measured by 4π proportional counter worked under elevated pressure.¹ Hereafter the 4π scintillation counter with plastic scintillators has been used for the same purpose.² In both cases the only radiation counted were K-X photons, or partly L-X, as Auger electrons were considerably absorbed at the source holder and at the source itself. Further progress in standardisation of E.C. nuclides relied upon the $4\pi(x, e)\text{-}\gamma$ coincidence method. Scintillation counters with a NaI/Tl crystal as well as ionisation chambers were also used with rather poor counting efficiency as Auger electrons were not counted, in these cases, at all.

Here the 4π liquid scintillation method has been used for counting the following E.C. nuclides: chromium-51, manganese-54, iron-55, zinc-65, yttrium-88 and iodine-125. Auger electrons of low energy can be counted in these cases together with K-X photons, and therefore all the advantages of liquid scintillation method for low energy β -emitters counting are valid.

THE COUNTING EQUIPMENT

The counting equipment used for E.C. nuclides counting was the same as for carbon-14 standardisation.³ Counting could be carried out in parallel and in the coincidence system. The scintillator used was PBD in toluene (8 g PBD/l 1 toluene) and 4 g PPO + 0.5 g POPOP/l 1 toluene with Triton X-100, for some experiments.

THE COUNTING METHOD

Sources to be counted were prepared in the counting vials, by dissolving a small amount of a radioactive solution in the scintillator, together with 1 ml of pure alcohol

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(except when Triton X-100 was added). As in the case of β -emitter counting, the anode characteristics for the parallel system and for the coincidence system were checked for each nuclide to choose the proper e.h.t. value as the working parameter.⁴ The anode characteristics for iron-55 are shown in Fig. 1. After choosing the proper e.h.t. value, all sources were counted with a variable discrimination level (from 48 to 175 mV) to obtain the discrimination curves and to extrapolate the counting rate to the zero level of

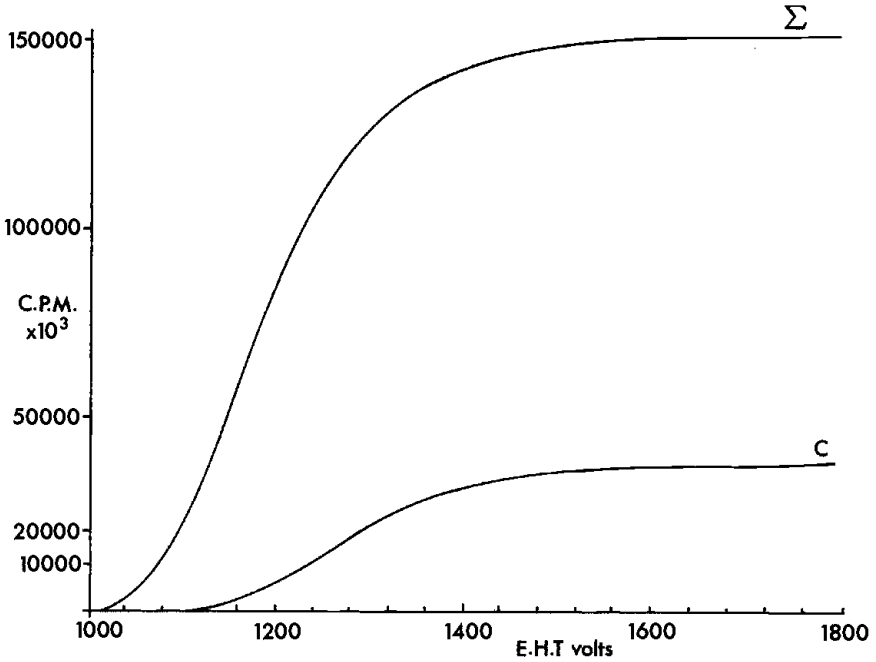


Fig. 1: The anode characteristics for iron-55. Σ : The parallel system. C: The coincidence system.

discrimination. The discrimination curve for iron-55 is shown in Fig. 2. All measurements were made with the additional quenching unit with $48\mu\text{s}$ of dead time. The counting temperature was -20°C .

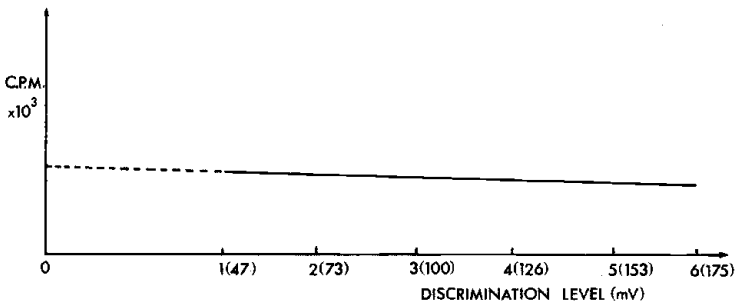


Fig. 2: The discrimination characteristic for iron-55.

THE COUNTING EFFICIENCIES

The list, with results, of counting efficiencies is given in Table 1. All measurements are for PBD in toluene scintillator with 1 ml of pure alcohol.

To obtain the counting efficiency the standard solutions, of the radionuclides in question, were used with radioactive concentration determined by the $4\pi(x,e)\text{-}\gamma$ coincidence method or 4π plastic scintillation counter. As it is shown in Table 1 the counting efficiency increased with the energy of radiation and seemed to be similar for K-X photons as for Auger-electron counting. To prove this observation the additional experiment was done, where only K-X of iron-55 photons were counted. The iron-55 source was prepared on transparent plastic foil, covered with another foil and placed into liquid scintillator. Auger-electrons were fully absorbed in the foils and only K-X photons were counted.

Taking K-X yield ($\frac{K-X}{N_0}$) into account the obtained counting efficiency was very close to that shown in Table 1.

THE QUENCHING EFFECT

To prove the possibility of application of the liquid scintillation method for activity measurements of E.C. nuclides, the quenching effect has been checked for the nuclides in questions. This was done by counting the standard samples with additional amounts of carrier of the same chemical composition. The quenching effect for manganese-54 is shown in Fig. 3.

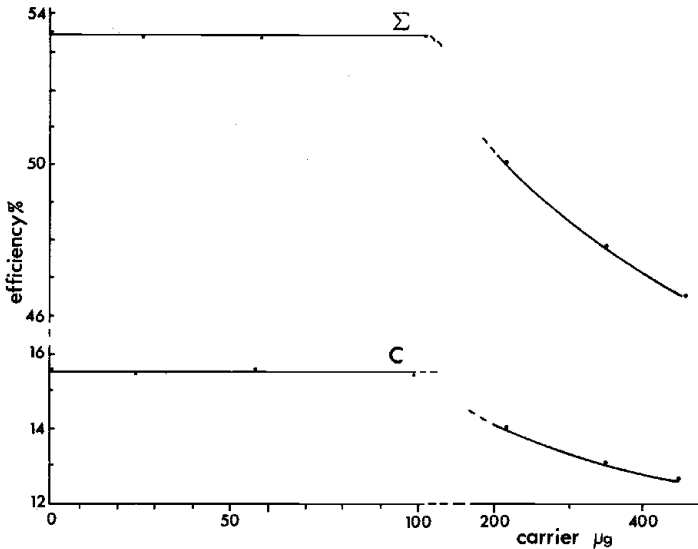


Fig. 3: The counting efficiency for manganese-54 and PBD in toluene as a function of the Manganese carrier. Σ : The parallel system. C: The coincidence system.

The counting efficiency was stable and the quenching effect was not observed up to $100\mu\text{g}$ of carrier. It should be stated that the amount of carrier in the normal counting sample is not more than several μg . Table 1 became an additional test for the quenching effect, as the results obtained did not depend upon the mass of the samples. It should be

Table 1. The counting efficiency with PBD in toluene as scintillator.

No of sample	Nuclide	K-X (keV)	Energy (keV)	Amount of samples	Mass of samples (g)	Chemical composition	Counting efficiency coinc. %	Supplier of stand. solut.	Error of stand. %	Counting error coinc. %	paral. %
1	chromium-51	4.95	325	4	0.01518 →0.02543	25 µg Cr as K ₂ CrO ₄ + 25 µg Cr as CrCl ₃ in 1 g 0.1 NHCl + 40 µg NaCl	14.9	R.C. Amersham	±1.4	±1.2	—
2	manganese-54	5.4	842	4	0.00640 →0.02595	25 µg Mn as Mn/NO ₃ / ₂ in 1 g 0.01 NHNO ₃ + 0.1% forma- line	15.5	R.C. Amersham	±2.8	±1.3	±2.0
3	iron-55	5.9	—	6	0.01470 →0.02656	25 µg Fe as FeCl ₃ in 0.1 N HNO ₃	16.0	IBJ Warsaw	±1.8	±1.4	±2.5
4	zinc-65	8.05	1115	4	0.00590 →0.03195	20 µg Zn as ZnCl ₂ in 1 g 0.1 N HCl	31.7	IBJ Warsaw	±1.1	±2.2	±1.6
5	yttrium-88	14.2	1836 898	5	0.013799 →0.050588	10 µg Y in 1 g 0.1 N HCl + 0.1% formaldehyde	39.6	IAEA Vienna	±2.0	±1.7	—
6	iodine-125	27.4	35.3	4	0.00536 →0.02099	25 µg I as KI + 50 µg Na ₂ S ₂ O ₃ in 1 g solution + 0.1% forma- line	50.3	R.C. Amersham	±2.7	±2.5	±3.5

also emphasised that the quenching effect was stronger for the coincidence system, than for the parallel one, as is shown by the counting ratio of the two counting systems in Fig. 4.

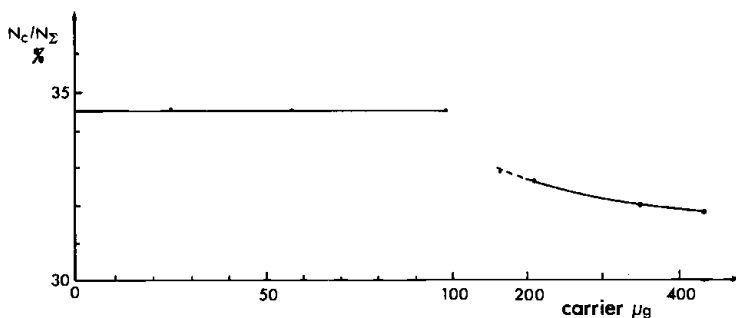


Fig. 4: The ratio of the coincidence system counting rate to the parallel system counting rate for manganese-54 (PBD in toluene scintillator) as a function of the carrier.

CONCLUSIONS

As it was pointed out, the liquid scintillation counting is a convenient method for activity measurements of E.C. nuclides. Although full counting efficiency was not obtained for any investigated nuclide, and the absolute method could not be realised, the obtained counting efficiencies were probably the highest from any possible counting method. The obtained counting efficiency for iron-55 can be compared with that for the scintillation counter with plastic scintillators,² where the counting sample was prepared in plastic foil and covered with another foil. The overall efficiency was lower at the case of plastic scintillators, as Auger electrons were completely absorbed in the foils, but full counting efficiency was obtained for K-X rays. This discrepancy was probably due to the light collection ability, which was much more effective for a point source and plastic scintillator sandwich, than for a distributed source at the liquid scintillator with an additional energy loss in wall effects.

An additional advantage of the 4π liquid scintillation method in the case of iron-55 is the possibility of activity measurement, independently from the yield factor (W_K) which is taken as a basis for activity calculation in the methods actually in use. The values for the yield factor (W_K) published by several authors show discrepancies of up to 10%. Providing that the counting efficiency for K-X photons and Auger-electrons are very close to each other, the counting efficiency for iron-55 can be obtained by interpolation of the values for other nuclides (chromium-51, manganese-54, cobalt-57) which can be standardised by the $4\pi(x,e)\text{-}\gamma$ coincidence method.

It should be also noticed that application of Triton X-100 was not successful in one investigation. The counting efficiency was generally lower (about 50% that of toluene scintillator) and the quenching effect investigation did not give reproducible results.

REFERENCES

- 1 R. A. Allen, *Metrology of Radionuclides*, International Atomic Energy Agency, Vienna, (1960) p. 343.

- 2 A. Tada and T. Radoszewski, *Standardisation of Radionuclides*, International Atomic Energy Agency, Vienna, (1967) p. 293.
- 3 T. Radoszewski, Chapter 2 of this book.
- 4 T. Radoszewski, *Nukleonika* 5, 361 (1960).

DISCUSSION

E. Langenschiedt: Which effects in your scintillation methods do you call 'wall effects'? Can you define them? Do they influence your extrapolation of the discriminator curve.

T. Radoszewski: The wall effect in standardisation is a geometrical concept and it is due to the loss of particles or photon energy in the wall instead of the scintillator. It could not be covered by the extrapolation curve—or, at least, only partly.

J. A. B. Gibson: In addition to wall effects there is a reduced efficiency at low energies due to ionization quenching when the rate of energy deposition (dE/dX) is high. The specific light output (dL/dX) is given by: $dL/dX = S \cdot dE/dX / (1 + kB \cdot dE/dX)$ where S is the scintillation efficiency and kB is a constant. The effect is shown in Fig. 6 in the discussion to Chapter 2.

T. Radoszewski: Yes, but still for α -emitters the wall effect would not have any influence on counting efficiency.

W. R. Greig: Is it possible to determine the counts due to iodine-125 and to tritium respectively when both are present in solution in a toluene PPO/POPOP system?

T. Radoszewski: I have not investigated this problem, but owing to the very poor resolution power for small energy in liquid scintillators, I think it will be difficult to distinguish the pulses from iodine-125 and tritium.

B. W. Fox: I can comment that to count iodine-125/tritium mixtures, the iodine-125 standard is counted in a gamma-counter and also in the tritium channel of a liquid scintillation counter. The tritium counts calculated from the instrument rates are subtracted from the total counts in the tritium channel to assess the tritium present. Also with Triton X-100 systems, it is the figure of merit which matters most, not absolute efficiency. What figure of merit can be achieved in this system?

T. Radoszewski: I was not interested in the figure of merit as I usually count very small amounts of solution (about 10 mg). I just measured to check the efficiency of counting for Triton X-100.

J. A. B. Gibson: Referring to iodine-125 and tritium counting, the resolution for the iodine-125 (i.e. X-ray at 27.4 keV) would be approximately 50% and thus for small amounts of tritium there will be a large contribution from iodine-125 in the tritium channel.

M. I. Krichevsky: In double label counting of isotopes with overlapping spectra it is best to allow maximum overlap in the various channels (without identity) rather than try to achieve maximum separation. Simultaneous equations are used to separate the counts due to each isotope in each channel and then compensated for quenching.