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The Use of the Cerenkov Effect in the Counting of β- and γ- Emitting Radionuclides

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In a transparent medium, β-radiation and the electrons which are released, under certain conditions, by the interaction of γ-radiation with the medium cause Cerenkov-radiation. The intensity of this Cerenkov-radiation was measured in water, glycerol and saturated sodium iodide solution in water. The counting efficiency, the sensitivity and the accuracy of the Cerenkov counting method for both β- and γ-emitting radionuclides in aqueous samples were compared with those associated with competing counting methods. It is concluded that Cerenkov counting has some distinct advantages over liquid scintillation counting and sodium iodide well crystal counting for routine measurements on large aqueous samples containing activities well above background.

INTRODUCTION

CERENKOV-radiation is emitted when a charged particle traverses a transparent medium (e.g. water) with a velocity exceeding that of light in the medium[1,2], i.e. when:

$$\beta n > 1 \tag{1}$$

where n is the refractive index of the medium and β the ratio of the velocity of the particle in the medium and that of light in vacuum. This means that for any medium there is a threshold velocity corresponding to a threshold energy[3]:

$$E_{\text{thr}}^{\text{med}} = 0{\cdot}511\{-1 + \sqrt{[1 + 1/(n^2 - 1)]}\}\ \text{MeV}. \tag{2}$$

Cerenkov-radiation has been used in the determination of the activity of some β-emitting radionuclides (i.e. ^{32}P). The main advantage of this detection method over liquid scintillation counting is the absence of chemical quenching problems. For the detection of γ-emitting radionuclides the potential possibilities of the Cerenkov effect have until now not been fully explored.

As a matter of fact, the yield of Cerenkov-radiation produced by secondary electrons originating from the interaction of γ-rays with water is poor; therefore detection of γ-radiation via the Cerenkov-radiation in water may be expected to be inefficient.

However, by adding a component with a high effective atomic number to the water, the number of released photo- and Compton electrons increases and so does the efficiency of the detection via the Cerenkov effect. A second efficiency-enhancing effect of this addition is the reduction of the threshold energy caused by the accompanying increase of the refractive index. These two effects were investigated by measuring the increase in Cerenkov-radiation in a saturated sodium iodide solution and glycerol with respect to water.

EXPERIMENTAL

Chemicals and materials

The media used for the Cerenkov measurements are:

distilled water:

$$n_D^{20} = 1{\cdot}330; \quad E_{\text{thr}} = 0{\cdot}264\ \text{MeV}.$$

a 95 wt.% aqueous solution of glycerol (Brocades p.a.):

$$n_D^{20} = 1{\cdot}449; \quad E_{\text{thr}} = 0{\cdot}195\ \text{MeV}.$$

a saturated aqueous solution of sodium iodide (Merck p.a.), to which a few drops of a 0·1 N solution of sodium thiosulfate were added:

$$n_D^{20} = 1{\cdot}490; \quad E_{\text{thr}} = 0{\cdot}178 \text{ MeV}.$$

The radionuclides used are specified in the first columns of Table 1.

Procedures

(1) *Cerenkov measurements in normal vials* with β-emitting nuclides in the three media and with γ- and β,γ-emitting nuclides in water were performed by adding to an Ekco glass counting vial 1 ml of the solution of the radionuclide and 19 ml of the medium.

A few mg of the corresponding inactive material was added in order to prevent drifting of the count rate due to adsorption to the walls of the vial. The count rates were measured with an Ekco liquid scintillation counting equipment consisting of the units N530G and N664B using 500-fold amplification, 6 V discriminator setting and a high voltage corresponding to the plateau in the count rate versus high voltage curve.

The count rates were corrected for background and are further referred to as C_1^w and C_1^m, in water and in the two other media, respectively. Overall-detection efficiencies in this counting procedure are referred to as Σ^w and Σ^m in water and in the other two media, respectively; detection efficiencies of the $\beta(\gamma)$ rays in this counting procedure are referred to as $\sigma_\beta^w(\sigma_\gamma^w)$ and $\sigma_\beta^m(\sigma_\gamma^m)$ in water and in the other two media, respectively.

(2) *Cerenkov measurements in special vials* with γ- and β,γ-emitting nuclides in the three media were performed with the aid of a specially designed vial (Fig. 1). To several identical plugs of the cover of the vial μC-amounts of the radionuclides were added.

The thickness of the bottom of the plugs was sufficient for the absorption of the β-radiation of the β,γ-emitting nuclides used.

TABLE 1. Detection efficiencies for aqueous solutions

Nuclide		Supplier* and code	E_β^{max} (MeV)	E_γ (MeV) (Φ)**	σ_β^w	σ_γ^w
β-emitting	^{185}W	1; NEZ-103	0·43		0·013	
	^{204}Tl	2;DRN-8100	0·77		0·083	
	^{32}P	2;DRN-1500	1·71		0·437	
	^{90}Y	1;NEZ-108	2·27		0·599	
	$^{144}Ce/^{144}Pr$	2;DRN-9001	2·98		0·612	
γ-emitting	^{51}Cr	2;DRN-2407		0·32		0·014
	^{85}Sr	1;NEZ-082		0·51		0·036
	^{54}Mn	1;NEZ-040		0·84		0·048
β,γ-emitting	^{131}I	2;DRN-5300		0·36		
	^{198}Au	2;DRN-7900		0·41		
	$^{137}Cs/^{137m}Ba$	2;DRN-5500		0·66		
	^{46}Sc	1;NEZ-076	0·36	0·89, 1·12	0·006	0·058
	^{60}Co	2;DRN-2703	0·31	1·17, 1·33	0·001	0·070
	^{88}Y	2; –	0·60	0·90, 1·85 (0·92),(0·08)	0·040	0·082

* 1 = New England Nuclear; 2 Philips Duphar.

** Φ (=relative intensity) is mentioned if different from 1·00

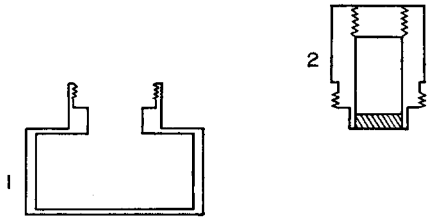


FIG. 1. The counting vial of procedure 2. (1) Vial (perspex); (2) demountable plug (perspex with aluminum cover and bottom).

Fifteen ml of the medium was placed in the vial; the count rates were measured and corrected for background and are further referred to as $C_2{}^w$ and $C_2{}^m$ in water and in the other two media, respectively.

Overall-detection efficiencies in this counting procedure are further referred to as $'\Sigma^w$ and $'\Sigma^m$, in water and in the other two media, respectively.

(3) *Geiger–Müller measurements* of the β-emitting nuclides were performed by transferring V (about 5×10^{-2}) ml of the solution of the radionuclide to a polyester-foil (thickness about 0·85 mg/cm^2), evaporating the drop to dryness and counting under strictly known geometrical conditions with an end-window Geiger–Müller detector (Philips PW 4107/01-18506) connected to a Philips counting equipment consisting of the units PW 4022, 4032 and 4052.

The count rates were corrected for background, coincidence losses and absorption in the air and the window and are further referred to as C_3. The geometric detection efficiency in this counting procedure was:

$$\eta = 0{\cdot}132.$$

(4) NaI-*well crystal measurements* of the γ- and β,γ-emitting nuclides were performed by transferring V (about 5×10^{-2}) ml of the solution of the radionuclide and 1 ml water to a centrifuge tube which was placed in the well of a NaI-scintillation crystal (Ekco N597). The count rates were measured with an Ekco scaler N530G, corrected for background and are further referred to as C_4.

The detection efficiency λ as a function of gamma-energy for a well-crystal of the same dimensions was taken from the data published by HOLMBERG *et al.*[4].

RESULTS

(1) Cerenkov measurements in water and Geiger–Müller measurements with the β-emitting nuclides were used to calculate $\sigma_\beta{}^w$ by:

$$\sigma_\beta{}^w \equiv \Sigma^w = \frac{C_1{}^w}{C_3} V \eta \tag{3}$$

(The results are given in Table 1 and Fig. 2.)

(2) Cerenkov measurements in water (procedure 1) and NaI-well measurements with the γ-emitting nuclides were used to calculate $\sigma_\gamma{}^w$ by:

$$\sigma_\gamma{}^w \equiv \Sigma^w = \frac{C_1{}^w}{C_4} V \lambda \tag{4}$$

(The results are given in Table 1 and Fig. 3.)

(3) Cerenkov measurements in water (procedure 1) and NaI-well measurements on some β,γ-emitting nuclides were used to calculate $\sigma_\gamma{}^w$. This was done for the nuclides ^{46}Sc and ^{60}Co by*:

$$\sigma_\gamma{}^w = 1 - \sqrt{\left(1 - \frac{\Sigma^w - \sigma_\beta{}^w}{1 - \sigma_\beta{}^w}\right)} \tag{5}$$

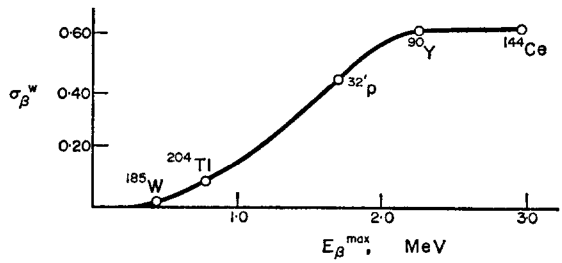


FIG. 2. The Cerenkov counting efficiency for bêta-radiation in water as a function of the maximum bêta-energy.

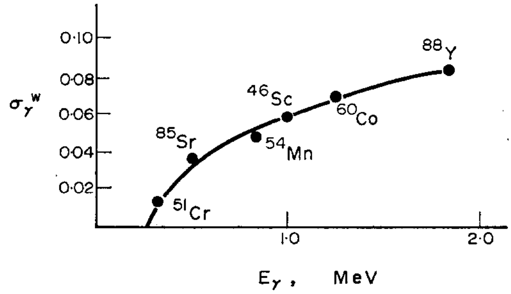


FIG. 3. The Cerenkov counting efficiency for gamma-radiation in water as a function of the gamma-energy.

* Derivation, see Appendix.

and for the nuclide ^{88}Y by:

$$\sigma_\gamma{}^w \equiv \sigma_{\gamma_2}{}^w = \frac{\Sigma^w - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{22}\sigma_\beta{}^w)}{1 - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{22}\sigma_\beta{}^w)} \qquad (6)$$

In both equations (5) and (6) Σ^w was calculated by:

$$\Sigma^w = \frac{C_1{}^w}{C_4} V\Lambda \qquad (7)$$

where Λ equals, for the nuclides ^{46}Sc and ^{60}Co:

$$\Lambda = \lambda_1 + (1 - \lambda_1)\lambda_2 \qquad (8)$$

and for the nuclide ^{88}Y:

$$\Lambda = \Phi_1(\lambda_1 + (1 - \lambda_1)\lambda_2) + \Phi_2\lambda_2 \qquad (9)$$

In these equations Φ denotes relative intensity and the indices of Φ, λ and γ refer to the different γ quanta emitted.* Values of $\sigma_\beta{}^w$ were taken by interpolation from Fig. 2.

(The results are given in Table 1 and Fig. 3.)

(4) Cerenkov measurements with the β-emitting nuclides in the three media were used to calculate $\Delta\sigma_\beta/\sigma_\beta{}^w$ by:

$$\frac{\Delta\sigma_\beta}{\sigma_\beta{}^w} \equiv \frac{\sigma_\beta{}^m - \sigma_\beta{}^w}{\sigma_\beta{}^w} = \frac{C_1{}^m - C_1{}^w}{C_1{}^w}$$

(The results are given in Fig. 4.)

(5) Cerenkov measurements with the γ- and β,γ-emitting nuclides in the three media (procedure 2) were used to calculate $\Delta\sigma_\gamma/\sigma_\gamma{}^w$ by:

$$\frac{\Delta\sigma_\gamma}{\sigma_\gamma{}^w} \equiv \frac{\sigma_\gamma{}^m - \sigma_\gamma{}^w}{\sigma_\gamma{}^w} \approx \frac{'\Sigma^m - '\Sigma^w}{'\Sigma^w} = \frac{C_2{}^m - C_2{}^w}{C_2{}^w}$$

(The results are given in Fig. 5a.)

DISCUSSION

β-emitting radionuclides

Figures 2 and 4 show that the Cerenkov counting efficiency for β-radiation increases, for the same medium, with increasing maximum β-energy and, for the same nuclide, with increasing refractive index of the medium. Both effects are explained by the fact that the fraction of electrons giving Cerenkov-radiation is increased, in the former by the increase of the maximum energy at constant threshold energy and in the latter by the decrease of the threshold energy at constant maximum energy.

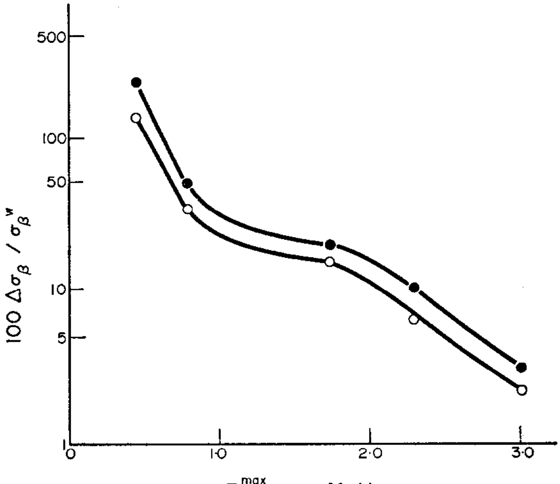


FIG. 4. The medium effect (in per cent) upon the Cerenkov counting efficiency for bêta-radiation as a function of the maximum bêta-energy (○: 95% glycerol, ●: saturated sodium iodide solution).

β, γ- and γ-emitting radionuclides

Figure 3 shows an increase of the Cerenkov counting efficiency for γ-radiation in water with increasing γ-energy. This effect can be explained, as above, by the fact that with increasing

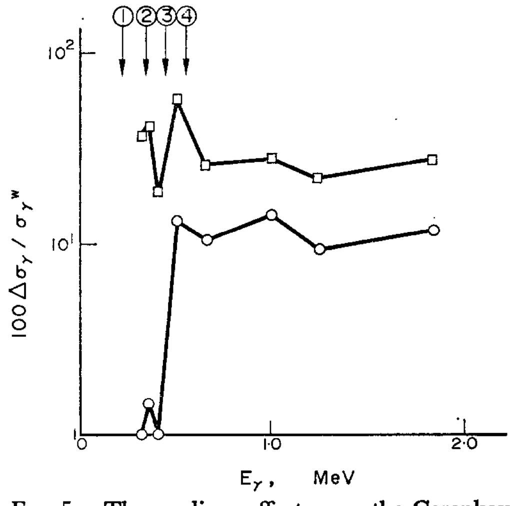


FIG. 5a. The medium effect upon the Cerenkov counting efficiency for gamma-radiation as a function of the gamma-energy. (○: 95% glycerol, □: saturated sodium iodide solution; the circled numbers refer to γ-energies corresponding with the situations depicted in Fig. 5b).

* See the Appendix for the derivation of equations (5) and (6).

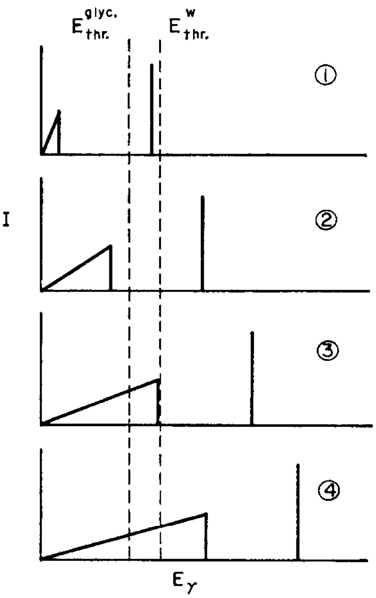


FIG. 5b. Schematical representation of the secondary electron spectrum at different gamma-energies.

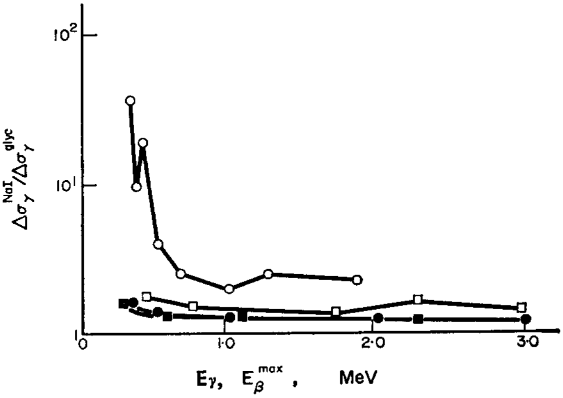


FIG. 6. The ratio of the medium effects exerted by saturated sodium iodide solution and 95% glycerol for β- and γ-radiation as a function of energy. (○: γ-radiation, experimental, ⊡: β-radiation, experimental, ●: β-radiation, theor., forbidden, ■: β-radiation, theor., allowed).

γ-energy at constant threshold-energy the relative number of (photo- and Compton-) electrons giving Cerenkov-radiation is increased.

The effect of the substitution of another medium for water on the Cerenkov counting efficiency for γ-radiation, at constant γ-energy, can be seen in Fig. 5a. In glycerol, this effect is caused only, as above, by the decrease of the threshold-energy, thereby increasing the fraction of Compton-electrons giving Cerenkov-radiation. However, in contrast with the effect on the Cerenkov counting efficiency for β-radiation (compare Fig. 4) this effect is practically zero at low γ-energy. This is explained by the discontinuous character of the spectrum of electrons released by the γ-radiation, as is schematically depicted in Fig. 5b.

In sodium iodide solution an additional effect is the increase of the ratio of photo- to Compton-electrons, resulting from the interaction with a medium of higher atomic number.

The effects are clearly demonstrated in Fig. 6, where the ratio of the medium effects exerted by the two investigated media on the Cerenkov counting efficiency for γ-radiation is plotted against the γ-energy. For comparison the analogous ratio for the Cerenkov counting efficiency for β-radiation is also plotted in this figure against the maximum β-energy, together with theoretical values of this ratio, computed by graphical integration of two typical β-distributions (of the allowed and the forbidden transition type) with boundaries corresponding with the threshold-energies in water and the two investigated media.

Comparison with other counting techniques

In the comparisons made in this section, we use data (detection efficiencies, background count rates, sample volumes) obtainable with our equipment. This is—cf. the experimental section—a one-photomultiplier tube liquid scintillation counter, which can be used for Cerenkov and liquid scintillation counting by placing a counting vial on top of the photomultiplier tube and which can be modified for γ-counting by mounting on top of the photomultiplier tube a sodium iodide well crystal having roughly the same dimensions as the counting vial in the liquid scintillation counting mode (the dimensions of the well may vary).

In Table 2 typical data for these three detection methods are given. It must be noticed that the comparisons hold for **aqueous** samples, since a value of 2 ml has been used for the liquid scintillation detection mode. This equals the volume of aqueous sample which can be mixed with 18 ml of non-aqueous scintillation medium.

TABLE 2. Data for different detection methods

Detection method	Sample volume (ml)	Background count rate (c.p.m.)
Liquid scintillation	2	150
Cerenkov	20	1500
NaI well crystal	1 and 5	700

In Fig. 7 the **counting-efficiency** for β-radiation of the Cerenkov detection method in water and saturated sodium iodide solution is compared with that of the liquid scintillation counting method. It can be seen that the liquid scintillation counting technique has the greatest counting efficiency over the entire energy range.

In Fig. 8 the **counting-efficiency** for γ-radiation of the Cerenkov detection method in water and saturated sodium iodide solution is compared with that of the sodium iodide well crystal counting method. Data for the latter method were taken from HOLMBERG *et al.*[4]. It can be seen that the detection efficiency of the sodium iodide well crystal technique is much better over the whole energy range.

One can define the **absolute sensitivity** of a counting method as the activity of a radionuclide giving a net count rate which equals the background count rate. As the highest detection efficiencies from the curves in Fig. 7 and 8

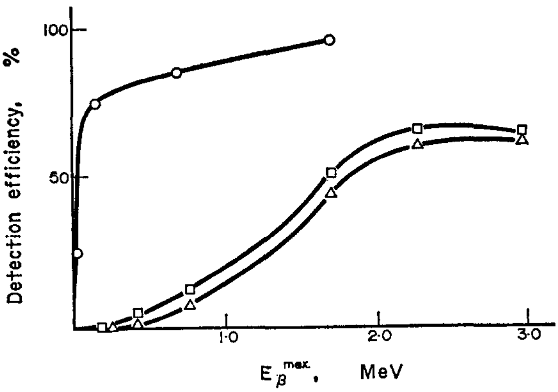


FIG. 7. The counting efficiency of various counting techniques for bêta-radiation as a function of the maximum bêta-energy. (⊙: liquid scintillation counting, △: Cerenkov counting in water, ⊡: Cerenkov counting in a saturated NaI-solution).

are associated with the counting techniques having the smallest background count rates (see Table 2) it can be concluded that the absolute sensitivity of the Cerenkov counting technique for both β- and γ-radiation is worse than that of the two competing techniques.

One can also define the **concentration sensitivity** of a counting method as the activity per ml of an (aqueous) radionuclide solution giving a net count rate (in the volume used for the measurement) which equals the background count rate. Values of this concentration sensitivity, obtained by dividing the

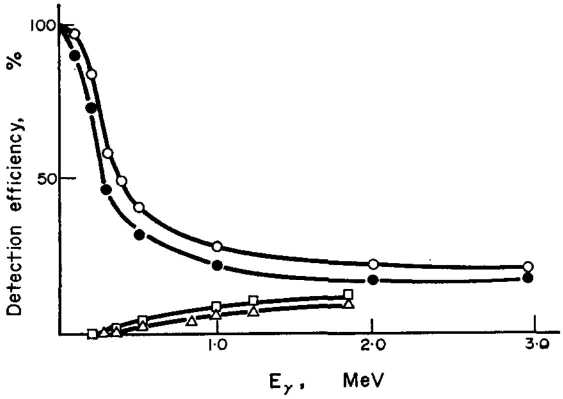


FIG. 8. The counting-efficiency of various counting techniques for gamma-radiation as a function of the γ-energy. (⊙: NaI(Tl)-Ekco N597-well crystal, sample-volume 1 ml, ●: NaI(Tl)-Ekco N597-well crystal, sample-volume 5 ml, △: Cerenkov counting in water, ⊡: Cerenkov counting in a saturated NaI-solution).

data for the absolute sensitivity by the corresponding data for the sample volume (Table 2), for β- and γ-radiation, are plotted in Figs. 9 and 10 against maximum β- and γ-energy, respectively.

It can be seen from Fig. 9 that the liquid scintillation detection method has a better concentration sensitivity than the Cerenkov detection method over the whole range of β-energies. From Fig. 10 it is clear that the concentration sensitivity of the Cerenkov counting method for γ-emitting radionuclides is comparable to that of the sodium iodide well crystal counting method (always for the conditions of this measurement) if the sample volume in the

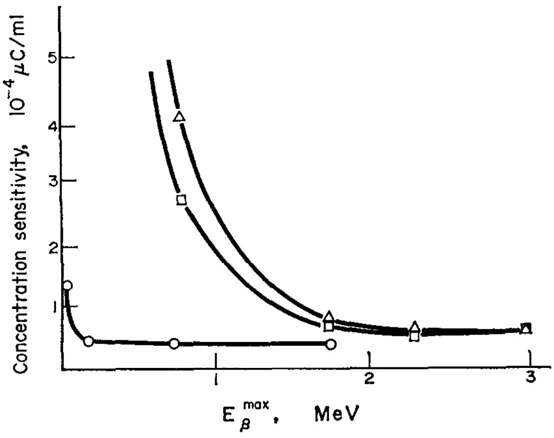


FIG. 9. The concentration sensitivity of various counting techniques for bêta-emitting radionuclides as a function of the maximum bêta-energy (notation as in Fig. 7).

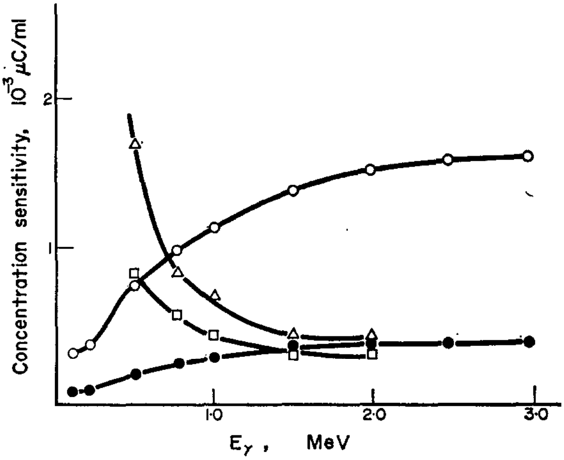


FIG. 10. The concentration sensitivity of various counting techniques for gamma-emitting radionuclides as a function of the gamma-energy (notation as in Fig. 8).

latter method is 5 ml and even better if the sample volume is 1 ml.

If one has to count many samples containing activities well above background the preset time counting mode is often preferable. In that mode, if sufficient quantities of the sample are available, the **relative standard deviation** of the net count rate is roughly inversely proportional to the square root of the product of detection efficiency and sample volume. In Figs. 11 and 12 these data are plotted against maximum β- and γ-energy, respectively, for the various detection methods compared.

As can be concluded from these figures, the statistical reproducibility is greater for the Cerenkov detection method for maximum β-energies and for γ-energies exceeding about 0·8–1·0 MeV.

Resuming, it can be stated that the Cerenkov detection method for β- and γ-emitting radionuclides in large aqueous samples containing activities well above background has the advantage of a greater statistical reproducibility than competing detection techniques in

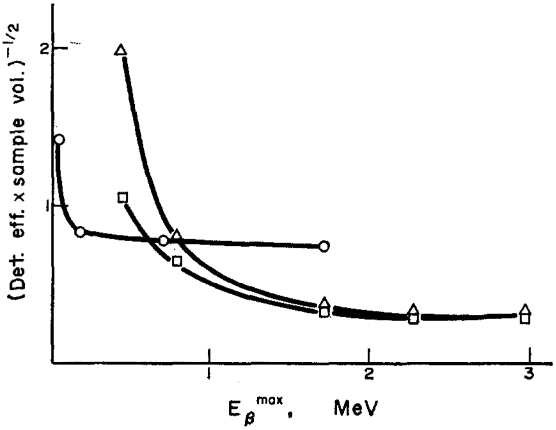


FIG. 11. A comparison of the statistical reproducibility of the various counting techniques for bêta-emitting radionuclides as a function of the maximum bêta-energy (notation as in Fig. 7).

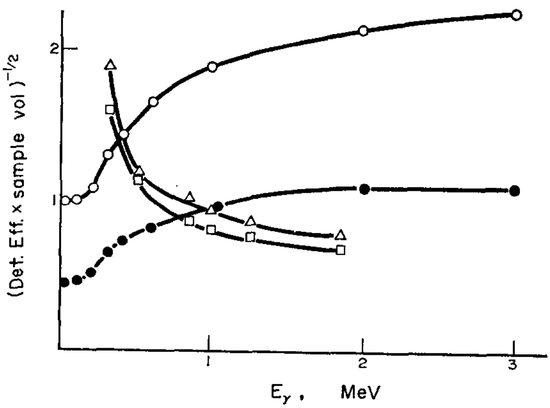


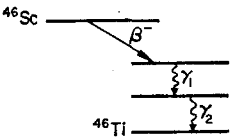
FIG. 12. A comparison of the statistical reproducibility of the various counting techniques for gamma-emitting radionuclides as a function of the gamma-energy (notation as in Fig. 8).

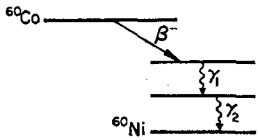
the preset time mode. (In the preset count mode, the advantage is in terms of reduced counting time). As has been referred to in the introduction, an additional advantage of Cerenkov detection of β-radiation over the liquid scintillation method is the absence of chemical quenching problems. Further, no expensive scintillation chemicals are required. Moreover, γ-detection by the Cerenkov detection method of β, γ-emitting radionuclides has the additional advantage over the sodium iodide well crystal method that the accompanying β-radiation substantially contributes to the detection efficiency (for ^{46}Sc, ^{60}Co and ^{88}Y Σ^w is about 2 times as large as $\sigma_\gamma{}^w$).

APPENDIX

Equations (5) and (6) are derived by considering the simplified disintegration diagrams of the radionuclides.

These are for ^{60}Co and ^{46}Sc as follows:





The overall Cerenkov detection efficiency in water (Σ^w) is equal to:

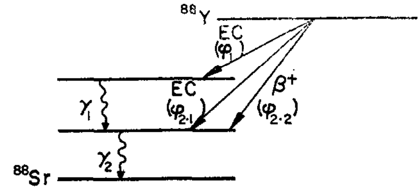
$$\Sigma^w = \sigma_\beta{}^w + (1 - \sigma_\beta{}^w)\sigma_{\gamma_1}{}^w + [1 - \sigma_\beta{}^w - (1 - \sigma_\beta{}^w)\sigma_{\gamma_1}{}^w]\sigma_{\gamma_2}{}^w$$

Assuming that $\sigma_{\gamma_1}{}^w \approx \sigma_{\gamma_2}{}^w = \sigma_\gamma{}^w$ this equation can be written as:

$$\Sigma^w = \sigma_\beta{}^w + (1 - \sigma_\beta{}^w)\sigma_\gamma{}^w + [(1 - \sigma_\beta{}^w) - (1 - \sigma_\beta{}^w)\sigma_\gamma{}^w]\sigma_\gamma{}^w.$$

$$\sigma_\gamma{}^w = 1 - \sqrt{\left[1 - \frac{\Sigma^w - \sigma_\beta{}^w}{1 - \sigma_\beta{}^w}\right]} \qquad (5)$$

The disintegration diagram of ^{88}Y is as follows:



In this case the overall Cerenkov detection efficiency is equal to:

$$\Sigma^w = \Phi_1[\sigma_{\gamma_1}{}^w + (1 - \sigma_{\gamma_1}{}^w)\sigma_{\gamma_2}{}^w] + \Phi_{2\cdot1}\sigma_{\gamma_2}{}^w + \Phi_{2\cdot2}[\sigma_\beta{}^w + (1 - \sigma_\beta{}^w)\sigma_{\gamma_2}{}^w]$$

$$\Sigma^w = \Phi_1\sigma_{\gamma_1}{}^w + \Phi_1\sigma_{\gamma_2}{}^w(1 - \sigma_{\gamma_1}{}^w) + \Phi_{2\cdot1}\sigma_{\gamma_2}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w + \Phi_{2\cdot2}\sigma_{\gamma_2}{}^w(1 - \sigma_\beta{}^w)$$

$$\Sigma^w - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w) = \sigma_{\gamma_2}{}^w[\Phi_1(1 - \sigma_{\gamma_1}{}^w) + \Phi_{2\cdot1} + \Phi_{2\cdot2}(1 - \sigma_\beta{}^w)]$$

$$\sigma_{\gamma_2}{}^w = \frac{\Sigma^w - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w)}{[\Phi_1(1 - \sigma_{\gamma_1}{}^w) + \Phi_{2\cdot1} + \Phi_{2\cdot2}(1 - \sigma_\beta{}^w)]} = \frac{\Sigma^w - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w)}{\Phi_1 - \Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot1} + \Phi_{2\cdot2} - \Phi_{2\cdot2}\sigma_\beta{}^w} = \frac{\Sigma^w - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w)}{1 - (\Phi_1\sigma_{\gamma_1}{}^w + \Phi_{2\cdot2}\sigma_\beta{}^w)} \qquad (6)$$

REFERENCES

1. Marshall J. *Phys. Rev.* **86,** 685 (1952).
2. Collins G. B. and Reiling V. G. *Phys. Rev.* **54,** 499 (1938).
3. Ross H. H. *Analyt. Chem.* **41,** 1260 (1969).
4. Holmberg P., Rieppo R. and Passi P. *Int. J. appl. Radiat. Isotopes* **23,** 115 (1972).
5. Elrich R. H. and Parker R. P. *Int. J. appl. Radiat. Isotopes* **19,** 263 (1968).