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Efficiency of 4π–Crystal–Scintillation Counting: 1. Experimental Technique and Results

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The technique of 4π-crystal-scintillation counting has been applied to the standardization of beta-emitting nuclides. Phototubes viewing an anthracene sandwiched source at 180° have been used with high-gain nonoverloading amplifiers in addition and in coincidence circuits to obtain high detection efficiencies. A precise shorted delay-line coincidence analyzer is described. The results of measurements made with P^{32}, Co^{60}, Sr^{90}-Y^{90}, I^{131}, and Tl^{204} are compared with those obtained by $4\pi\beta$-gas-proportional counting.

1. Introduction

The method of $4\pi\beta$-gas-proportional counting has been established for some 6 years now for the primary standardization of beta-emitting nuclides at the National Bureau of Standards [1, 2, 3].[2] The method of 4π-liquid-scintillation counting has also recently been investigated at the Royal Cancer Hospital in London [4]. It was therefore considered to be of interest to initiate experiments to determine the efficiency of 4π-crystal-scintillation counting and its applicability to primary beta standardization.

These measurements have now been concluded and show that the method of 4π-crystal-scintillation counting, although not as generally applicable as $4\pi\beta$-gas-proportional counting, can be used for medium and high-energy beta-emitting nuclides. It is the purpose of this paper to report briefly the experimental results obtained for the benefit of other interested workers in the field.

2. Experimental Procedure

The crystal scintillation counters used in these experiments consisted of sandwiches formed by two transparent anthracene crystals each approximately 1 in. in diameter and ranging from ⅛ to ¼ in. in thickness. Two Dumont 6292 multiplier phototubes, in contact with the backfaces of the anthracene crystals, were used with high-gain nonoverloading amplifiers to detect light pulses occurring within the crystals. One of the experimental arrangements is shown in figure 1. The phototubes were inside a light-tight refrigerator, with the lowered temperatures being used to reduce the random thermal tube noise. The entire sandwich is surrounded by an aluminum cylinder for complete light collection. In this connection it was found that a 2-in. diameter cylinder gave slightly better light collection than one of only slightly larger diameter than that of the crystals, presumably because the former enabled reflected light to reach the phototube directly without being retransmitted through the crystals or scattered by them.

Negative high voltage was supplied to the phototubes so that their anodes could be connected directly to the cathode-follower input grids. Voltages as high as −1,400 v were used and electronic amplifier gains of 40,000 were, in some cases, found necessary.

The radioactive material was generally introduced into the sandwich by depositing an aliquot of the active solution directly onto the top face of the lower crystal. The deposit was allowed to dry and then covered by the second crystal to form the sandwich. Where special techniques were used to precipitate the active material the sources were prepared on thin Formvar-polystyrene laminated films ($10\mu g/cm^2$) and then placed on the crystal face.

The pulses from the phototubes arising from the light pulses in the crystal sandwich were counted in two ways, namely in addition and in coincidence. In the former method the separate phototube-cathode-follower output pulses were added together and then fed into a single amplifier and counted. It was found that a single tube and hemispherical reflector arrangement was not as efficient as the two-tube addition method. In the coincidence method the output pulses were fed into separate amplifiers, thence into a coincidence analyzer of short ($1\mu sec$) resolving time and the coincidences counted.

Each of these methods, two-tube addition and coincidence, has advantages and disadvantages. In the coincidence method the random noise pulses originating in each phototube will not be counted except for those few accidental coincidences arising during the finite resolving time of the coincidence circuit. However, it is necessary that a true light pulse originating inside the sandwich be "seen" by *both* phototubes. This stringent requirement is absent in the addition method so that the sensitivity to very small light pulses is higher by at least a factor of 2 as compared with the coincidence method. On the other hand, the addition method suffers from the limitation of excessive phototube noise at the high tube gains sometimes required. Because the phototube noise is usually not reproducible to better than ±5 to 10 percent the uncertainty in the knowledge of the true disintegration rate can become

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[2] Figures in brackets indicate the literature references at the end of this paper.

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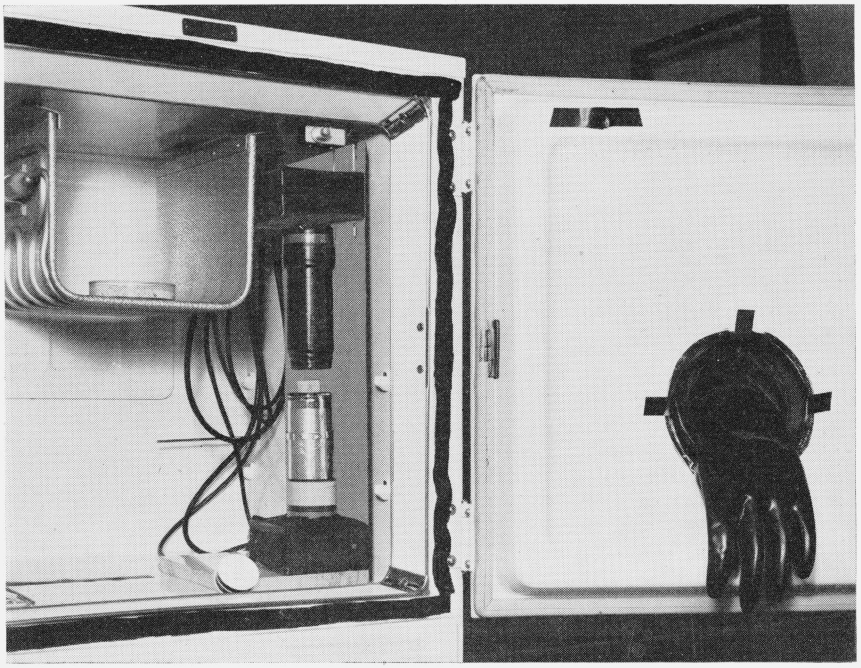

FIGURE 1. *Anthracene crystal sandwich and multiplier phototubes assembly in refrigerator.*

appreciable. A noise counting rate of 2,000 counts per second in each channel would, in the addition method, give rise to approximately 4,000 counts per second to be subtracted from the total observed counting rate. However, in the coincidence method, with a resolving time of 1 μsec, the increase in coincidence counting-rate observed would be only 4 counts per second. Because this number is a small fraction of the "true" counting rate any uncertainty in the knowledge of the exact noise background will be reduced in this ratio.

The coincidence method presupposes that at least one photoelectron is emitted from *each* photocathode in order that there be a finite probability of detecting a coincidence. Assuming a 5 percent photocathode photoelectric efficiency and a geometrical light-collecting efficiency of 100 percent, one would require a minimum of 40 photons (20 photons to each phototube) to give rise to a coincidence count. However, in order to insure that *both* photocathodes receive at least 20 photons with a 99 percent probability it would require a minimum of 68 initial photons. With the further assumption of a quantum efficiency of 65 ev/photon for anthracene [5] the effective beta-ray cutoff energy would correspond to 4.4 kev. In the addition method, however, either phototube would have a threshold of 20 photons. This requirement is not as stringent as in the coincidence method and the effective cutoff energy would be 2.2 kev. In the $4\pi\beta$-gas-proportional counting method the effective energy cutoff is estimated to be only 0.2 kev.

For high-energy beta emitters such as P^{32} and Sr^{90}-Y^{90} the preponderance of the emitted beta rays have energies greater than 2.2 kev. Further, the light pulses are rather large so that the phototube gain need not be high. Thus the noise is low and the addition method is satisfactory. However, for the lower-energy beta emitters the phototube gain must be increased, increasing the random tube noise and necessitating the use of the coincidence method.

A precision shorted delay-line coincidence analyzer was designed for the coincidence measurements and is shown schematically in figure 2. The unit accepts positive pulses from the nonoverloading amplifiers, shapes them and feeds the shaped pulses into a biased mixing stage. The rise and decay times of the square-shaped pulses are 0.06 μsec. The resolving time is variable by means of a shorted delay-line and the resolving time is independent of counting rate up to 5,000 counts per second in each channel. The single channel outputs record only those pulses that have been accepted by the input discriminator and which are on their way to the coincidence mixing stage.

3. Results and Discussion

A good fraction of the data was taken at room temperature. Operation at temperatures below 0° C considerably improved the precision of the tube-noise measurements and permitted the use of higher phototube voltages than was previously feasible. To illustrate the improvement due to lowered temperatures and consequent lowered tube noise, as well as the difference between the coincidence method and the addition method, several relative efficiency curves have been reproduced in figure 3. The efficiencies relative to $4\pi\beta$-gas-proportional counting are plotted as functions of phototube voltage for both the addition and the coincidence

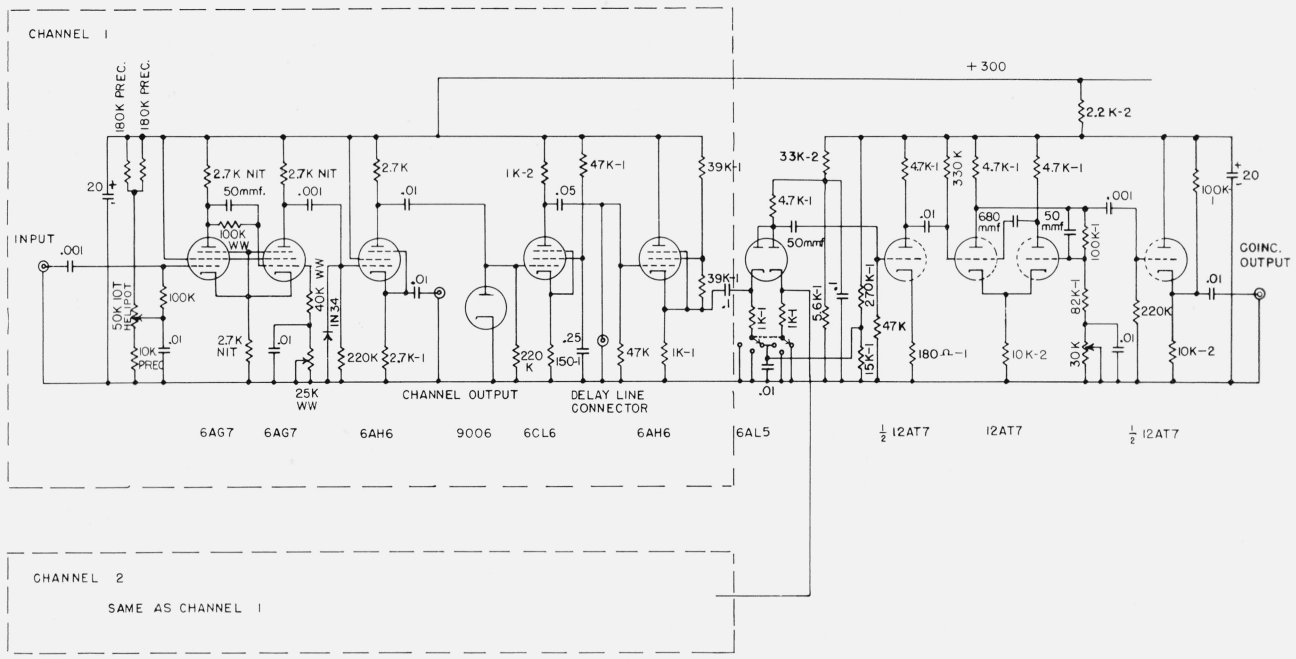

FIGURE 2. *Shorted delay-line coincidence-analyzer circuit.*

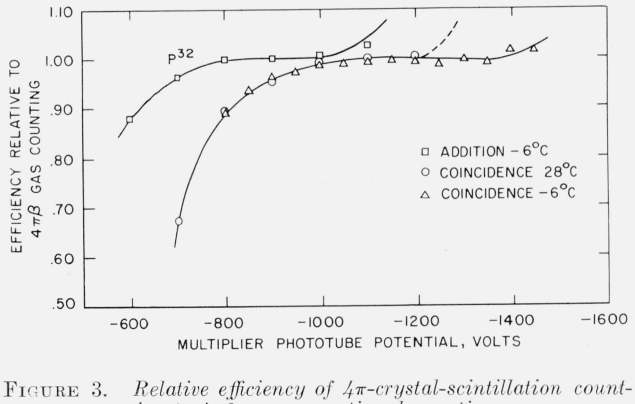

FIGURE 3. *Relative efficiency of 4π-crystal-scintillation counting to 4πβ-gas-proportional counting.*

methods at 28° C and at −6° C. In each case the usable plateau is considerably lengthened at the lowered temperature. These data were obtained for a high-energy beta emitter, namely P^{32}. For low-energy beta emitters the plateau shrinks to a very small region of phototube voltage and the improvement due to lowered temperatures is even more striking. For example, Tl^{204} ($E_{\beta max}=0.67$ Mev) exhibited a plateau only at the lowered temperature for the addition method. At room temperature the thermal-noise background at the phototube gains required was so large and erratic as to completely mask any plateau. Because of its better noise-discrimination characteristics the coincidence method was used for all of the lower-energy beta emitters, at room temperatures and at the lowered temperatures. It is particularly in these cases that the corrections for dead-time and coincidence resolving time become important. In a separate paper [6] both approximate and rigorous derivations for these counting losses are given and their range of applicability is discussed. Because it is shown that the approximate derivation can be employed under the present experimental conditions, only these corrections will be summarized here.

In channel I, consisting of phototube I, amplifier I and discriminator and gate I, a counting rate $N(\mathrm{I})$ is observed with

$$N(\mathrm{I})=N_n'+N_{tc}. \tag{1}$$

N_n' is the observed thermal tube noise of phototube I and N_{tc} is the observed "true" counting rate due to the source. Similarly in channel II for phototube II viewing the opposite side of the sandwich

$$N(\mathrm{II})=N_n''+N_{tc}. \tag{2}$$

The dead-time losses are due to the introduction of an electronic gate τ_g ($\tau_{g\mathrm{I}}=\tau_{g\mathrm{II}}$) whose duration is in all cases longer than the resolving time of the non-overloading amplifier. The use of this fixed gate insures a precise correction for dead-time as the amplifier resolving time is dependent upon the degree of overload or gain. To a first approximation the fractional dead-time loss due to noise and "true" counts in both channels is given by

$$L=\tau_g\,(N(\mathrm{I})+N(\mathrm{II})-N_{tc}+N_n'N_n''\tau_g), \tag{3}$$

remembering that N_{tc} occurs in *both* channels simultaneously and should be corrected for only *once*. Again to a first approximation the observed coincidence rate N_c is related to N_{tc} by

$$N_c=N_{tc}+2\tau_c N_n'N_n'', \tag{4}$$

where τ_c is the resolving time of the coincidence analyzer; the last term in eq (4) being the accidental coincidence rate due to random noise. From eq (3) and (4) one obtains the total disintegration rate, N_t, as

$$N_t = \frac{N_c - 2\tau_c N_n' N_n''}{1 - \tau_g[N(\mathrm{I}) + N(\mathrm{II}) - (N_c - 2\tau_c N_n' N_n'') + N_n' N_n'' \tau_g]}. \quad (5)$$

In a blank experiment, i. e., no source present,

$$(N_c)_b = (N_{tc})_b + 2\tau_c (N_n' N_n'')_b \quad (6)$$

$(N_{tc})_b$ arises from cosmic rays, possible contamination of the crystals, and fluorescence in the glass of the phototubes. In actual practice $(N_{tc})_b \ll (N(I,\mathrm{II}))_b$ so that with very little error N_n' in eq (5) and (6) can be set equal to $(N(\mathrm{I}))_b$. Likewise $N'' \simeq (N(\mathrm{II}))_b$. The disintegration rate of the source N_0 is then given to within a few tenths of a percent by

$$N_0 = N_t - (N_{tc})_b \quad (7)$$

Figure 4 shows the extent of these corrections in the cases of Tl^{204} and Co^{60}. The dashed curves represent the observed values of $N_c - (N_c)_b$ as functions of phototube voltage for both isotopes at 28° C and at −6° C. The solid curves, representing these data corrected by means of eq (5), (6), and (7), give N_0, the actual source disintegration rate. Due to the high noise counting rates at room temperature the corrections required were appreciably higher than those for the lower temperature, making it impracticable to continue any measurements at room temperature past −1,200 v.

Table 1 gives a summary of data obtained by means of the 4π-crystal-scintillation technique for the nuclides P^{32}, Co^{60}, $Sr^{90}-Y^{90}$, I^{131}, and Tl^{204}, compared with measurements made in the $4\pi\beta$-gas-proportional counter.

This comparison points up a serious limitation of

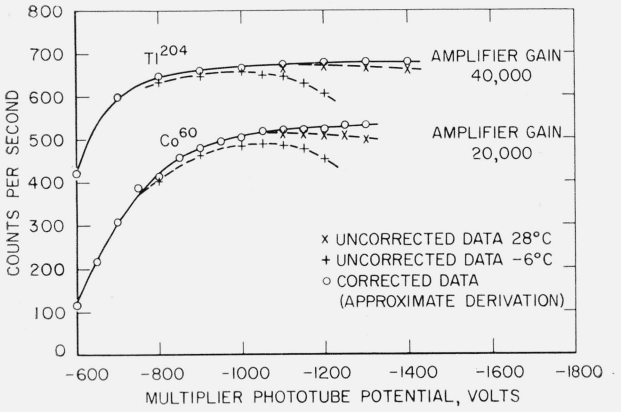

FIGURE 4. *Uncorrected data and corrected values of the absolute disintegration rates for sources of cobalt-60 and thallium-204.*

the 4π-crystal technique for low-energy beta emitters. Because the minimum pulse observable is that caused by a single photoelectron being emitted from the photocathode there is a definite energy cutoff that will depend upon (a) the intrinsic photon efficiency of the crystal, (b) the transmission of the crystal to its own light, (c) the external light-collecting geometry, and (d) the photoelectric efficiency of the photocathode. The variations in observed efficiency for Co^{60} shown in table 1 are probably due to combinations of these factors.

It is probably the case that for low-energy beta particles the photon efficiency of crystal anthracene will be lower than 1 photon per 65 ev [5]. If one assumes an average efficiency of 95 percent for the 4π sandwich-crystal method in the case of Co^{60} it is possible to make some estimate of this value for anthracene for low-energy beta particles. Ninety-five-percent coincidence efficiency corresponds to 97.5-percent single-channel efficiency or a 2.5-percent loss of low-energy beta particles. As roughly 2.5 percent of the Co^{60} beta spectrum is below 5 kev in energy this would correspond to a minimum energy cutoff of 5 kev. Again assuming a 5-percent photoelectric efficiency for the phototube photocathode one obtains a photon efficiency for anthracene of the order of 1 photon per 120 ev in the range 0 to 5 kev. This is not an unreasonable figure, for in this range the specific ionization of electrons is a very rapidly rising function in the direction of decreasing energy.

TABLE 1. *Summary of data for various nuclides*

Nuclide	$4\pi\beta$-gas counting	4π-crystal counting	Ratio $\frac{\text{Crystal}}{\text{Gas}}$
	dps	*dps*	
P^{32}	274 ±1	275 ±3	1.00
	627 ±3	628 ±3	1.00
Co^{60}	619 ±3	607 ±5	0.98
	609 ±2	597 ±10	.98
	573 ±2	530 ±5	.93
$Sr^{90}-Y^{90}$	880 ±4	884 ±4	1.00
	840 ±4	832 ±8	0.99
I^{131}	741 ±3	736 ±5	.99
	277 ±1	273 ±2	.99
Tl^{204}	371 ±1	373 ±3	1.00
	681 ±2	680 ±3	1.00

4. Conclusions

The 4π-crystal-scintillation technique is suitable for the standardization of medium and high-energy beta emitters and in this energy range gives results that agree quite well with results obtained with the $4\pi\beta$-gas-proportional counter. One possible advantage of the former method is the higher density of anthracene relative to the counter gas and the consequently higher efficiency for the detection of X-rays from electron-capturing nuclides. This may prove useful in the standardization of electron-capturing nuclides.

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WASHINGTON, July 3, 1956.

5. References

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