The Absolute Standardization of Radioisotopes by 4 π Counting

H. H. Seliger and L. Cavallo

The absolute counting of beta particles, using 4 π proportional flow counters, is described. This method of absolute counting has been adopted in the Radioactivity Section of the National Bureau of Standards for the primary calibration of radioisotope solution standards. Pillbox, cylindrical, and spherical 4 π flow counters have been used in the measurements, and excellent results have been obtained for each. A simple method of source mounting has been developed, and a formula is derived that determines the absorption in the source mounting regardless of knowledge of the thickness of the mounting or energy spectrum of the beta rays. Results are given of intercomparisons of P^{32} and I^{131} with other laboratories in the United States, Canada, and Great Britain that employ different types of absolute counting techniques. The applicability of the 4 π counting method to the standardization of practically all beta emitting radiosotopes is discussed.

I. Introduction

4 π proportional flow counters have been used at the National Bureau of Standards for the determination of absolute disintegration rates of radioisotopes since September 1949.[1] The principle of 2 π and 4 π counting is well known to workers in the field of radioactivity. However, several improvements have been made in the measurement technique, and since the method has not been covered previously in any detail it was felt that a description of the technique used would be of practical interest. This is particularly true in view of the excellent agreement that has been obtained with other published absolute counting methods.[2 3 4 5]

II. Technique

To eliminate the difficulties associated with Geiger counters and still retain somewhat their pulse limiting property, the 4 π counter is operated in the region of limited proportionality.[6 7] Figure 1 shows the disassembled cylindrical and pillbox 4 π counters.[8 9] The insulators are of Teflon. The stainless steel cylindrical counter was designed with the length equal to the diameter, so that the minimum path length for a particle emitted in any direction from the center of the counter would be as close as possible to the average path length. A 0.001-in. conducting diaphragm, also visible in figure 1, fits between the two halves and serves to divide the cylinder into two equal hemicylinders so that the top and bottom halves function independently. The source is mounted on the polystyrene face of a laminated film of Formvar and polystyrene[10] approximately 40 $\mu g/cm^2$ in thickness, and this thin film is cemented over the diaphragm center hole.

99 Mole percent of methane has been used as a flow gas in all counters and has been found to be preferable to an Argon-Methane flow mixture. Both the Bell Jordan linear amplifier[11] and the Brookhaven nonoverloading amplifier[12] have been used.

Two comparison tests are made on all readings taken with the 4 π counter. First integral discrimination curves are plotted for various electronic gains, keeping the counter voltage constant, and second, these discrimination curves are plotted for various counter voltages or gas gains, keeping the electronic gain constant. In both cases if the discrimination curves are flat, one can assume that all primary beta particles are being detected.

The curves shown in figures 2 and 3 illustrate the completeness of the electron collection and also the method of extrapolation to find the true disintegration rate. Even though the discrimination curves have been found in practice to be flat, it can be assumed that some primary beta particles are not detected, their number being too small to contribute appreciably to the slope of the curve. The gas amplification in this region is 10^3, and the electronic gain is 5000. The capacity of the counter circuit is approximately 10^{-11} farad. Therefore a primary beta ray must create in the neighborhood of 12 ion pairs in order for its pulse to be detected, assuming that an output pulse must be at least 10 volts in height. This would preclude from measurement all primary beta rays with energies less than 400 ev[13] which is a negligible fraction in the cases considered.

[1] Grateful acknowledgement is made to C. J. Borkowski of the ORNL, who suggested this method at a meeting of the Subcommittee on Beta and Gamma Ray Measurements and Standards of the Committee on Nuclear Science of the National Research Council in June 1949.

[2] An account of the theory and experimental application is given by L. H. Gray, British J. Radiology **22**, 677 (1949).

[3] A spherical collecting geometry is described by R. K. Clark, Columbia University, Rad. Res. Lab., N. Y. O.—1506 (March 15, 1950).

[4] A parallel plate collecting geometry is described by W. Gross and G. Failla, Phys. Rev. **79**, 209 (1950).

[5] T. B. Novey, Rev. Sci. Instr. **21**, 280 (1950).

[6] S. A. Korff, Electrons and nuclear counters (D. Van Nostrand Co., 1946).

[7] Rossi and Staub, Ionization chambers and counters (McGraw-Hill, 1949).

[8] Rossi and Staub, Ionization chambers and counters, p. 130 (McGraw-Hill, 1949), illustrate a 4 π counter used for α particles.

[9] A diagram of a 4 π counter is shown in a review article on beta counting of J. L. Putnam, United Kingdom, AERE-NR-318 (Feb. 24, 1949.)

[10] L. M. Fry and R. T. Overman, Atomic Energy Com. Doc. 1800 (Jan. 30, 1948).

[11] P. R. Bell and W. H. Jordan, Rev. Sci. Instr. **18**, 703 (1947).

[12] W. Bernstein, H. G. Brewer, Jr., and W. Rubinsin, Nucleonics **6**, 39 (1950).

[13] This agrees roughly with the calculations of R. Ballentine, private communication.

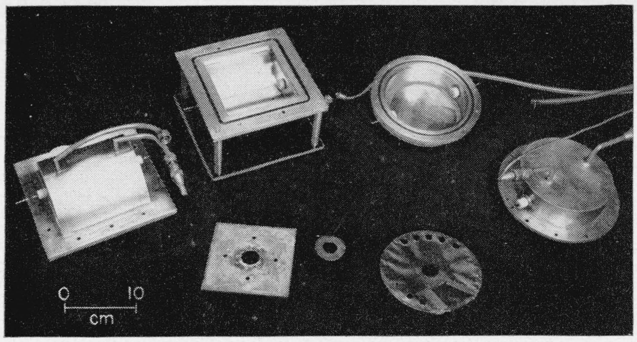

FIGURE 1. *Disassembled cylindrical and pillbox 4 π counters.*

The "0" ring Teflon insulators are clearly visible. In the foreground are the diaphragms and the 0.001 in. aluminum source mounting ring. The relative scale is given by the [centimeter rule] in the [center].

III. Source Mounting

Experiments have been conducted to determine the amount of absorption, if any, due to the film between the source and the lower half of the 4π counter. The number of particles counted by the top half of the counter connected separately will be

$$N_{\text{top}}=\frac{N_0}{2}\,[1+B_F+(1-\tau)B_w(b)], \qquad (1)$$

where N_0 is the true disintegration rate of the source, B_F is the percentage backscattering from the film, τ is the fractional absorption in the film, and $B_w(b)$ is the percentage backscattering due to the walls in the bottom half. The number of particles counted by the bottom half connected separately will be

$$N_{\text{bottom}}=\frac{N_0}{2}\,[(1-\tau)+(1+B_F)B_W(t)]. \qquad (2)$$

From symmetry considerations $B_W(t)=B_W(b)=B_W$.

The factor B_F can be neglected when the film is thin and of low atomic number, so that (1) becomes

$$N_{\text{top}}=\frac{N_0}{2}\,[1+B_W-\tau B_W]. \qquad (3)$$

In the bottom half, again assuming $B_F=0$, one obtains

$$N_{\text{bottom}}=\frac{N_0}{2}\,[(1-\tau)+B_W]. \qquad (4)$$

Putting this in the form $y=ax+b$ gives

$$N_{\text{bottom}}=\frac{-N_0}{2}\,\tau+\frac{N_0}{2}\,(1+B_W). \qquad (5)$$

With the thin films under consideration it can be assumed that the absorption is directly proportional to the film thickness. Equation (5) can be used to determine the absorption correction graphically. A more direct method of determining τ can be deduced from eq (3) and (4).

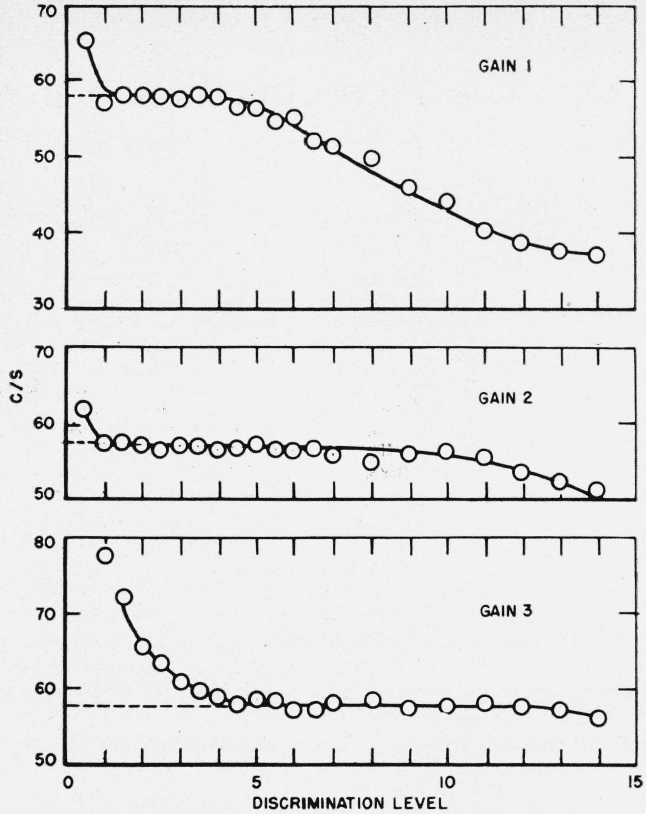

FIGURE 2. *Counting rate as a function of discrimination level for increasing steps in amplifier gain.*

The source is P^{32} in the pillbox 4 π counter. The voltage on the counter was held constant during all runs. Gain 2 is twice that of gain 1, and gain 3 is 4 times that of gain 1.

$$N_{\text{top}}-N_{\text{bottom}}=\frac{N_0}{2}\,\tau\,(1-B_W). \qquad (6)$$

The actual counting rate observed with top and bottom halves connected together is

$$N_{tb}=N_0\,(1-\tau/2) \qquad (7)$$

so that (6) becomes, if one lets $N_{\text{top}}-N_{\text{bottom}}=\Delta$

$$\tau=\frac{N_{\text{top}}-N_{\text{bottom}}}{N_{tb}-N_{\text{bottom}}}=\frac{\Delta}{N_{tb}-N_b}, \qquad (8)$$

and similarly

$$B_W=\frac{N_{\text{top}}+N_{\text{bottom}}}{N_{tb}}-1. \qquad (9)$$

The absolute counting rate is then obtained by substituting (8) into

$$N_0=\frac{N_{tb}}{1-\tau/2}. \qquad (10)$$

Thus by taking three different readings of the same source on a single film it is possible to determine the absorption by the film. This proves extremely useful for low energy beta particles.

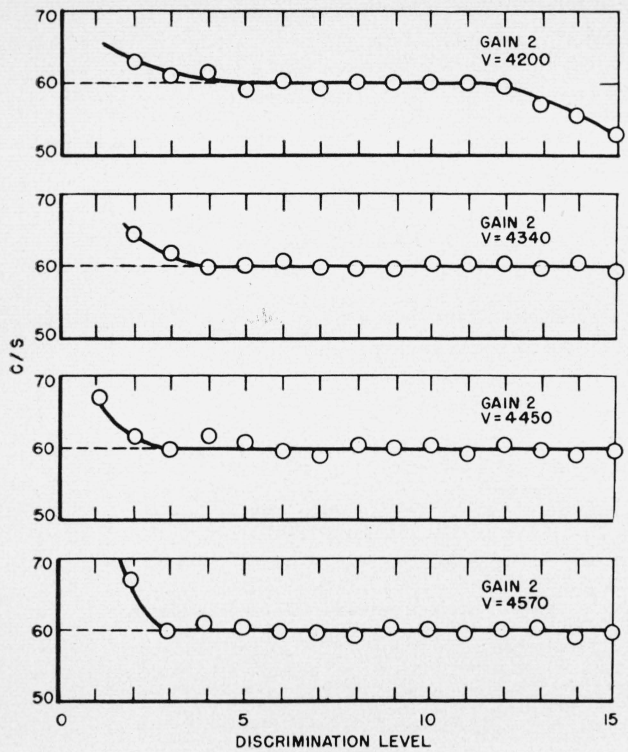

FIGURE 3. *Counting rate as a function of discrimination level for increasing steps in voltage.*

The source is I^{131} in the cylindrical 4 π counter. The electronic gain was held constant, and discrimination curves were run for various voltages on the counter.

Experiments have been performed with P^{32}, I^{131}, and CO^{60} using film thickness in multiples of approximately 40 $\mu g/cm^2$, and excellent agreement has been obtained between eq (5) and (9). As an example of the corrections necessary for absorption, for a film of 40 $\mu g/cm^2$ thickness $\tau=0$ for P^{32}, $\tau=0.01$ for I^{131}, and $\tau=0.05$ for Co^{60}. In the cases of I^{131}, Co^{60}, and C^{14} polystyrene films as low as 10 $\mu g/cm^2$ in thickness have been used. More complete data are now being taken for Co^{60}, C^{14}, and Na^{22}.

Previously when thin films were used as source mounts it was thought necessary to sputter a thin conducting layer of gold on both sides of the film so as not to disburb the collecting fields. However, if the area of the dielectric film is made small compared to the total area of the conducting diaphragm these collecting fields will be perturbed only slightly with no loss in efficiency. All results shown for the cylinder and spherical counters have been obtained using nonconducting plastic films, and table 1 shows the agreement for P^{32} between the cylinder counter in which such thin films were used as source mountings and the pillbox counter in which the sources were mounted on a conducting film of aluminum.

IV. Source Thickness

Equation (10) previously derived is valid, provided there is no self-absorption in the source itself. In any case the effect of self-absorption in a source of the same thickness as the mounting film is not as serious as the absorption in the film. The source is an evaporation covering a small area, and therefore there is a slowing down of electrons rather than complete stopping, as in the case of the film where some of the emitted electrons can have a long path length. Since the specific ionization curve has a maximum at approximately 7,000 ev, this slowing down usually adds to the total ionization. The effect of self-absorption will be more important in the cases of Co^{60}, C^{14}, and S^{35}.

V. Results and Conclusions

P^{32} and I^{131} sources have been calibrated in all three types of 4 π counters. The internal consistency of repeated measurements in all cases is within 1 percent. The results of some determinations made using the three types of counters are shown in table 1.

It is felt that the cylindrical 4π counter has slightly better collecting fields than either the sphere or the pillbox. This does not affect in any way the absolute counting of the beta particles, but is evidenced by a slightly longer voltage plateau for the cylinder. However, the spherical counter is by far the most economical, the total cost including modifications and mounting stand being approximately \$80.00.[14]

The cylindrical 4π counter is at present being used for all standardizations.

The results of a series of intercomparisons of P^{32} and I^{131} solutions originally calibrated at the National Bureau of Standards and distributed to other laboratories are given in table 2.

The technique described above is applicable to any beta emitting radioisotope, regardless of the complexity of the disintegration scheme. Isomeric states or gaseous isotopes may cause difficulties. However all effects due to gamma-ray line spectra, gamma-ray conversion electrons, or electrons produced in the gas or the walls of the counter by gamma rays only add to the total ionization per disintegration and are counted as one ionization pulse. A high specific activity source is not required, since such low counting rates can be recorded that water dilution is practically always possible. Most important, the method is self-consistent. Any improper mounting or positioning of the source is shown up immediately by a large value for τ. Any other malfunctioning will not result in flat discrimination curves. There are no further meter or condenser calibrations necessary. Equation (10) gives the true disintegration rate regardless of changes in temperature, humidity, pressure inside the chamber, or voltage.

The 4 π counter method is at present being used to calibrate sources of C^{14}, Na^{22}, and Co^{60}. As soon as practicable S^{35}, Na^{24}, Au^{198}, and Sr^{90} will be calibrated.

---

[14] Hemispheres are available from Nuclear Measurements Corp., 3339 Central Ave., Indianapolis, Ind.

Table 1. *Results of comparison measurements made of* P^{32} *and* I^{131} *in all three types of* 4π *counters*

[The errors indicated are statistical standard deviations]

Isotope and date of distribution	Counting rate	Type of counter	Type of amplifier	Result
	Counts/sec			*dps/ml*$\times 10^{-3}$
I^{131} 6/8/50	Less than 100	Pillbox	Bell Jordan	300±2
	do	Cylinder	do	298±1
P^{32} 9/14/50	do	do	do	239±0.63
	do	do	Brookhaven	241±0.40
	do	Sphere	Bell Jordan	240±0.24
	Approximately 900	Cylinder	Brookhaven	239±1.0

Table 2. *Results of independent intercomparisons of* P^{32} *and* I^{131} *made by other researchers using absolute counting techniques*

The errors, except for Dr. Hawkings' figures, are estimated inaccuracies in absolute value

Isotope and date of distribution	Activity dps/ml$\times 10^{-3}$				
	NBS 4π counter	L. H. Gray ionization chamber	R. C. Hawkins 4π counter [a]	Oak Ridge defined solid angle	G. Failla parallel plate chamber
I^{131} 12/13/49	250±7.5	248±6.2			
P^{32} 2/27/50	327±9.8		322±1.4	317	
I^{131} 6/8/50	299±9.0	300±1.2	304±1.7		
P^{32} 9/14/50	240±7.0	229±5	243±1	235±5	241±7

[a] R. C. Hawkings, Can. Nat. Research Coun., Chalk River, Canada, uses a spherical 4π counter similar to the NBS counter for his determinations.

The authors are indebted to R. C. Hawkings for his cooperation in the intercomparisons of the P^{32} and I^{131} solution standards, and to L. H. Gray, Hammersmith Hospital, London, Eng., G. Failla, Columbia University School of Medicine, E. I. Wyatt and J. A. Cox of the Oak Ridge National Laboratory for their interest and cooperation. Thanks is also given to L. Costrell of the Nucleonics Instrumentation Section for the construction of the electronic equipment, for his valuable assistance in setting up the pillbox 4π counter in September of 1949, and for his continued interest.

Washington, December 18, 1950.