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Refinements in Radioactive Standardization by 4π Beta Counting

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The corrections due to backscattering and film absorption for radioactive sources in 4π proportional flow counters have been investigated. The low-field effect arising from the use of thin, but nonconducting, Formvar-polystyrene films has also been investigated, and the results have been compared with those obtained in experiments carried out with such supporting films rendered conducting by the evaporation thereon of an approximately 15-microgram-per-square-centimeter layer of gold. Sandwiches of such conducting films have been used to determine the absorption due to the supporting film. It has been shown that the simpler nonconducting source-mounting film technique may still be used to calibrate radioactive sources to within the limits of accuracy currently in demand.

1. Introduction

In an attempt to eliminate some of the uncertainties of 4π counting, namely, those due to backscatteing and absorption by the source holder, Seliger and Cavallo [1] [1] adopted a method employing very thin plastic films on which to mount the sources and derived a simple relationship for determining N_0, the true disintegration rate of a radioactive source. This derivation was based, however, on the assumption that for a very thin film of low atomic number, the backscattering from the film could be neglected. A somewhat more rigorous derivation of the relevant relations than is given in the previous paper shows that, within the limits of error currently set for radioactive standardization, this assumption is, in fact, adequate.

In addition, in a 4π counter in which the collecting electrodes in the 2π hemispheres are of the same polarity and separated by a nonconducting film there must exist, in the proximity of the nonconducting film, a region around which there will be a low accelerating field that will be vanishingly small at a given point. The extent of the correction for this latter effect can not readily be calculated because it depends not only on the shape of the field and the exact position of the source in the field but also upon the proportion of β particles from the source that has too low an energy to penetrate into regions of higher field.

Experiments have therefore been carried out during the past year to investigate these effects and to determine the ultimate limitations of the method of 4π counting.

2. Effect of Backscattering

In the previous paper [1] it was assumed that the fractional backscattering from the film, β_f, could be neglected. But if this approximation, that β_f may be neglected, be left until the end of the derivation, it is possible to estimate the error arising from this assumption.

If, adopting the nomenclature of the previous paper [1], we assume that N_0 is the true disintegration rate of the radioactive source, that τ is the true fractional absorption in the film, and that N_{tb} is the combined counting rate for both hemispheres, then, without any assumption, N_{tb} is equal to N_0, less the number of particles absorbed per second in the source holding film. As the number of articles absorbed per second is $(N_0/2)\tau$, we have

$$N_{tb}=N_0-\frac{N_0}{2}\tau, \tag{1}$$

or, as in the previous paper,

$$N_0=\frac{N_{tb}}{1-\tau/2}. \tag{2}$$

If, as previously, N_t and N_b are, respectively, the separate counting rates in the top and bottom hemispheres, and if we also assume that β_f and β_f' are respectively the fractional backscattering from the film mount and from the combined film and aluminum partition between the two hemispheres and τ' is the combined fractional absorption of the film and partition, then we have

$$N_t=\frac{N_0}{2}\{1+\beta_f+\beta_w(1-\tau-\beta_f)(1-\tau'-\beta_f')\}, \tag{3}$$

where β_w is the fractional backscattering from the walls and the gas in either hemisphere.
Similarly,

$$N_b=\frac{N_0}{2}\{1-\beta_f-\tau+\beta_w(1+\beta_f)(1-\tau'-\beta_f')\}. \tag{4}$$

Thus from (1) and (4), and from (3) and (4)

$$N_{tb}-N_b=\frac{N_0}{2}\{(1+\beta_f)(1-\beta_w(1-\tau'-\beta_f'))\}, \tag{5}$$

and

$$N_t-N_b=\frac{N_0}{2}\{(2\beta_f+\tau)[1-\beta_w(1-\tau'-\beta_f')]\}. \tag{6}$$

Hence from (5) and (6)

$$\frac{N_t-N_b}{N_{tb}-N_b}=\frac{2\beta_f+\tau}{1+\beta_f}. \tag{7}$$

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

Because β_f and τ are both small compared with unity, it is possible, for the purpose of estimating the relative contributions of β_f and τ, to neglect β_f in the denominator on the right-hand side of eq (7). It is also found experimentally that the left-hand side is never greater than about 0.04. Thus, if in the extreme case, we assume that β_f is zero, there would be a correction due to τ in eq (2) amounting to 2 percent. If, on the other hand, we assume that β_f is of the same order of magnitude as τ, then, from eq (7), we have $3\tau \approx 0.04$, for the highest value of the left-hand side of eq (7), and the resultant correction for τ in eq (2) will be about 0.7 percent. The uncertainty introduced in neglecting β_f will, therefore, never exceed about 1.5 percent. This is well within the original limits of accuracy claimed for this method.

The type of backscattering referred to in this paper is not due to the diffusion of particles in the mounting film, as the film is quite thin, but to multiple scattering through small angles of particles originally incident at small angles with the plane of the film mount. This "side-scattering" has been shown [2] to be relatively independent of Z, so that for films of the thicknesses used in the previous paper [1] β_f for an evaporated gold film would be roughly equal to β_f for a Formvar-polystyrene film.

3. Experimental Methods

The approach to the problem has been by three different methods of experiment. In all these, the spherical type of 4π counter, already described [1], has been used. In the first place the existence of the low-field effect was demonstrated by means of a series of experiments in which I^{131} sources were deposited on Formvar-polystyrene nonconducting films supported on aluminum diaphragms having circular apertures of varying diameter. By varying the diameter of the aperture from 13 to 3 mm, the apparent value of N_0 was found to increase by approximately 3 percent. However, with the practical limitations of source preparation and with the necessity for keeping absorption by the diaphragm at a minimum, there will always be a region of low field near the center of a source, however small the diameter of the aperture, as long as there is a nonconducting film present. One can never be certain that the low-field effect is completely eliminated by this method. It has also been shown that the effect depends on the position of the source relative to the two fine wire loops forming the anodes of the two 2π hemispheres. Thus, if the source happens to be displaced toward one loop or the other, the counting loss tends to be lower in that hemisphere because the region of low fields for moderately large source-mount apertures will tend to remain at the center of the 4π counter. Using the second experimental approach to be described, it was found in one instance with I^{131} that by raising the source about ⅛ in., the low-field correction to N_t fell from 5.5 to 3.1 percent.

The second experimental method adopted is that which we have termed the "mirror image" experiment. In this experiment, in order to determine the low-field effect on the counts in one of the 2π hemispheres, the electrode in the other hemisphere has been maintained at an equal but opposite potential. By this means the lines of electric force will pass directly through the nonconducting source holder and the counting rate in the one hemisphere of the counter would be exactly the same as if the supporting film had been conducting. Thus by taking counts in one, and then the other hemisphere with the electrode in the opposite hemisphere first positive and then negative, the percentage loss in counts due to the low-field effect can be directly calculated for both sides of the source. It is not possible to put a negative voltage, of say 3,000 v, on one fine wire anode of the 4π counter and a positive voltage of 3,000 v on the other on account of electrical break-down due to field emission and corona discharge. However, for the purpose of determining the low-field correction, it is sufficient to substitute as the negative electrode a small sphere of about the same diameter as the wire loop to draw "lines of force" through the source and to remove the region of low field. The insertion of the sphere on its Teflon insulation in place of one of the anode wire loops can be achieved without disturbing the position of the source relative to the other loop and the appropriate correction determined. The steps in this experiment are illustrated in figure 1, where the nomenclature is the same as in the previous paper [1].

From experiments (a) and (b) represented in figure 1, the low field correction for N_b can be calculated; and from experiments (d) and (e), that for N_t. The actual correction necessary, however, is that to the combined counting rate N_{tb}. Because the corrections for N_t and N_b are usually of the same order, it will be sufficiently accurate to take the mean of the separate corrections for N_t and N_b for the correction to N_{tb}.

The third avenue of investigation consisted of evaporating extremely thin films of gold, of the order of 10 $\mu g/cm^2$, on to the Formvar-polystyrene films. An evaporation unit was built capable of operating at pressures of 5×10^{-5} mm Hg, at which very satisfactory gold-on-Formvar-polystyrene films were obtained. With these films as supports, sources of Co^{60}, I^{131}, and P^{32} were standardized by 4π counting. On account of the low energy of the β particles

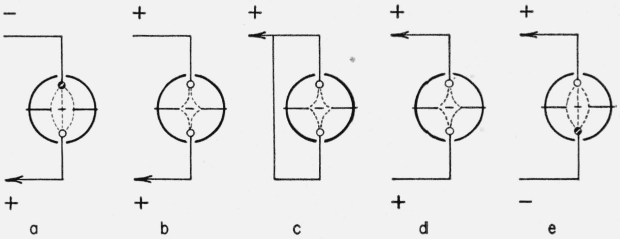

FIGURE 1. *Mirror image experiment.*

a, N_b determined with top sphere negative; b, N_b determined with top loop positive; c, N_{tb} determined; d, N_t determined with bottom loop positive; and e, N_t determined with bottom sphere negative.

With the nomenclature adopted, it is also usual for the source to be on the upper side of the supporting film so that, in general, $N_t > N_b$. The arrows indicate the hemisphere being counted.

emitted in the case of Co^{60} extreme precautions, such as the boiling of all glassware in detergents and then in conductivity water immediately prior to use in order to inhibit hydrogen ion exchange with the silica in the glass, had to be taken to reduce aggregation of solids and consequent self-absorption in the source itself. The thickness of the gold films was controlled by resistance measurements, a film on a control microscope slide 5 cm long and 1 cm wide having a resistance of about 60 to 100 megohms for a density of 15 $\mu g/cm^2$. They were adjudged to be of sufficient thickness by varying the thickness of gold and by the fact that on carrying out a mirror-image experiment on a conducting film-source mount, using Co^{60}, the correction to N_t was found to be zero.

In the experiments with the gold-Formvar-polystyrene films the fractional absorption due to the film was measured directly by carrying out the 4π measurements first with the source supported on a single gold-Formvar-polystyrene film and then sandwiched between two such films.

Here again the true disintegration rate of the source N_0 is readily obtained in terms of the combined counting rate N'_{tb}, for the sandwich, τ and β_f.

Suppose that we first obtain N'_{tb} for a single gold-Formvar-polystyrene supporting film with the source deposited on the gold. Equation (2) then gives N_0, provided we know τ, which is now the fractional absorption due to the laminated film. To obtain τ, we carry out a further experiment in which a similar film is placed on top of the source to form a sandwich, the film being so thin that it intimately adheres to the first. We assume that τ and β_f are the same for both films forming the sandwich, the gold having been evaporated from the same source at the same distance and at the same time. To obtain N_0 in terms of N'_{tb} we need merely to determine the number of particles totally absorbed in the sandwich. From figure 2 it is clear that those particles emerging into the top hemisphere directly will lose $(N_0/2)\tau$ by absorption. Particles emerging into the top hemisphere after being backscattered once from the bottom film will lose a total $(N_0/2)\beta_f\tau$ by absorption where β_f are the number backscattered from the film. Particles emerging into the top hemisphere after being backscattered twice will have lost $(N_0/2)\beta_f^2\tau$ by absorption and so on. Thus considering only one hemisphere, the total number absorbed will be represented by the infinite series:

$$\frac{N_0}{2}\tau(1+\beta_f+\beta_f^2+\ldots)=\frac{N_0\tau}{2(1+\beta_f)}. \tag{8}$$

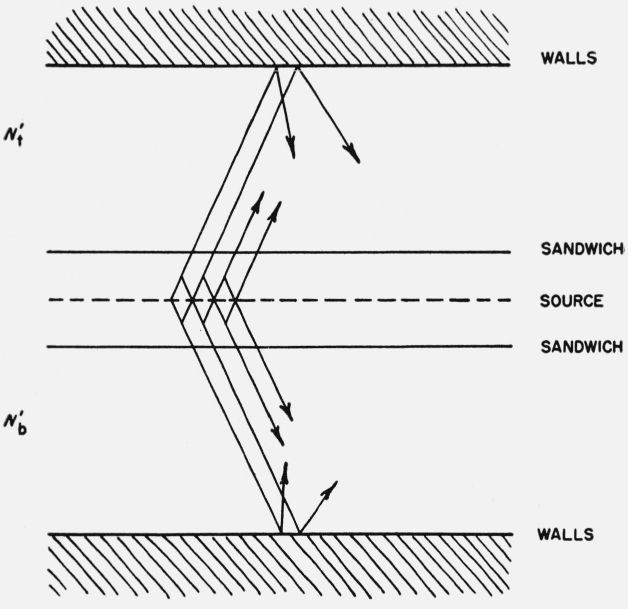

Figure 2.

Considering the contribution to both hemispheres, the total number absorbed will be double this. Thus without any assumption regarding the magnitude of β_f we have

$$N'_{tb}=N_0-\frac{N_0\tau}{1-\beta_f}$$

or

$$N_0=\frac{N'_{tb}}{1-\frac{\tau}{1-\beta_f}} \tag{9}$$

Here again β_f and τ are both small compared with unity, so that the effect on τ, which is already a relatively small correcting term, of neglecting β_f compared with unity will be in the second order of small correcting terms. We may, therefore, write

$$N_0=\frac{N'_{tb}}{1-\tau}, \tag{10}$$

and from (2) and (10) we immediately derive τ, the fractional absorption of the gold-Formvar-polystyrene film, as

$$\frac{\tau}{2}=\frac{N_{tb}-N'_{tb}}{2N_{tb}-N'_{tb}}. \tag{11}$$

From the foregoing analysis it is seen that in the case of the sandwich experiment, the effect of neglecting β_f is a second order effect and as such is much less serious in eq (9) than in eq (7). However, it is unlikely, using a nonconducting film and eq (7), even in the worst case of Co^{60}, to introduce an error greater than 1.5 percent. Moreover, by using the relatively good approximations for a gold-Formvar-polystyrene single film and a gold-Formvar-polystyrene sandwich, respectively

$$\frac{N_t-N_b}{N_{tb}-N_b}=2\beta_f+\tau, \tag{12}$$

and

$$\frac{N_{tb}-N'_{tb}}{2N_{tb}-N'_{tb}}=\frac{\tau}{2} \tag{13}$$

one should be able to obtain quite a good idea as to the value of β_f for such a gold-Formvar-polystyrene film. In the case of Co^{60}, using gold-Formvar-polystyrene films, we have found that $\tau \sim 0.05$ and $\beta_f \sim 0.03$.

In the present series of experiments we have compared the results obtained by the procedures of the previous paper [1] using a single nonconducting film, but introducing a correction for the low-field effect by means of the mirror-image experiment, with the results obtained using the conducting film sandwich technique.

4. Results

In table 1 the results obtained in the standardization of solutions of Co^{60}, I^{131}, and P^{32} are summarized. All Co^{60}, I^{131}, and P^{32} sources are derived from the same master solutions of Co^{60}, I^{131}, and P^{32}, in terms of 1 ml of which the final activities are given.

In the cases of Co^{60} and I^{131} a 2-percent correction has been made for self-absorption by the source. This correction is an estimated one based on average aggregate sizes determined by shadow-electron micrographs and high-power optical microscopic examinations of precipitated I^{131} and Co^{60} sources. These precipitated sources were shown to consist of clusters of solid with considerable open space in between. Thus, since addition of solid in the form of carrier could, within certain limits, presumably fill in the open spaces, it is not, as was thought previously, possible to extrapolate to zero solids in the source.

The Co^{60} solutions referred to in tables 1 and 2 have also been measured at the National Bureau of Standards by gamma-gamma coincidence counting and by R. W. Hayward, using both beta-gamma and gamma-gamma coincidence counting; and in Canada by R. C. Hawkings of Atomic Energy of Canada, Ltd. The results of this intercomparison are summarized in table 2.

TABLE 1. *Activities of* Co^{60} *as of May 7, 1952,* I^{131} *as of 8 a. m. on January 15, 1953, and* P^{32} *as of 8 a. m. on May 17, 1953, determined by 4π counting*

Source	Nonconducting film, no low-field correction	Nonconducting film, mirror image correction to—		Nonconducting film with mirror image correction	Conducting film, gold-Formvar-polystyrene
		N_t	N_b		
	dps/ml	*Percent*	*Percent*	*dps/ml*	*dps/ml*
381–7 (Co^{60})	277×10^5	5.2	3.4	289×10^5	------
381–X[a] (Co^{60})	------	------	------	------	290×10^5
386–101 (I^{131})	242×10^3	5.2	2.9	249×10^3	------
386–105 (I^{131})	234×10^3	4.3	3.0	244×10^3	------
386–X[a] (I^{131})	------	------	------	------	251×10^3
407–1A[4] (P^{32})	71.2×10^5	1.2	0.6	72.2×10^5	------
407–X[a] (P^{32})	------	------	------	------	[b](71.7×10^5)

[a] Average results for a large number of sources. The nonconducting film supports were all 17mm in diameter.

[b] This value was actually the mean of a number of values obtained with sources on nonconducting Formvar-polystyrene films but only 8 mm in diameter. For this diameter it had been shown, by comparison with conducting film source mounts, that the low-field correction was negligible for P^{32}.

TABLE 2. *Summary of standardization results obtained for* Co^{60}; *zero time, May 7, 1952*

Co^{60}				
Laboratory	4π counter, conducting film	4π counter, mirror image	β-γ coincidence	γ-γ coincidence
	mC/ml	*mC/ml*	*mC/ml*	*mC/ml*
National Bureau of Standards	0.785	0.782	0.787	0.790 .800
Atomic Energy of Canada, Ltd	--------	--------	.790	--------

5. Summary

Not only from the internal consistency of the two methods of 4π counting described, but also from the excellent agreement with the results obtained from coincidence counting at the National Bureau of Standards and at other laboratories, it would appear that with due care, 4π counting techniques, using either conducting or nonconducting films, are justified for such low-energy β-emitters as Co^{60}. Satisfactory results can be obtained with P^{32} without adopting any special precautions to eliminate self-absorption or the low-field effect. In the case of Co^{60}, however, the loss due to self-absorption, without extreme precautions as to source preparation has in some cases been found to be as high as 10 percent.

All primary calibrations by 4π β counting at the National Bureau of Standards are now made using as source mounts 15 μg/cm² conducting gold films evaporated on 20μg/cm² Formvar-polystyrene supports; film absorption corrections are made by the sandwich technique.

We gratefully acknowledge the helpful cooperation of R. C. Hawkings of Atomic Energy of Canada, Ltd. We also thank R. W. Hayward for an excellent set of β-γ and γ-γ coincidence measurements on Co^{60}, and L. Cavallo, S. V. Culpepper, and G. W. Randel for their valuable assistance in making the 4π counter measurements on I^{131}, Co^{60}, and P^{32}.

6. References

[1] H. H. Seliger and L. Cavallo, The absolute standardization of radioisotopes by 4π counting, NBS J. Research **47**, 41 (1951) RP2226.

[2] H. H. Seliger, Phys. Rev **88.**, 408 (1952).

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