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COUNTING METHODS FOR THE ASSAY OF RADIOACTIVE SAMPLES*

by

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TABLE OF CONTENTS

	<u>Page</u>
INTRODUCTION.	3
GENERAL CONSIDERATIONS.	3
BETA COUNTING.	6
End-window Counters	6
I (detection probability).	7
G (geometry).	8
f_W (factor for air and counter-window absorption)	9
f_A (factor for scattering effect of air)	10
f_C (factor for effect of sample covering).	11
f_B (factor for effect of backscattering).	12
f_H (factor for the effect of the housing).	14
f_S (self-absorption and self-scattering factor).	14
Suggested Techniques for End-window Counting.	16
Absolute Beta Counting	16
Relative Counting	18
2π Counting	18
4π Counting	20
Internal Gas Counting	23
External Gas Counting.	24
Coincidence Counting	24
Beta Counting of Liquid Samples	28
Beta Scintillation Counting.	28
ALPHA COUNTING.	31
Geometry Factor	31
Scattering.	31
Backscattering.	31
Absorption	32
Self-absorption	32
Plateaus	33
Beta Pulse Pile-up.	34
Counting Standards	34
Resolution Losses	34
Absolute Counting.	34
Relative Counting.	35
GAMMA COUNTING	36
Relative Counting.	36
Absolute Gamma Counting	36
Scintillation Counting - Pulse Height Analysis.	37

TABLE OF CONTENTS

	<u>Page</u>
ACKNOWLEDGMENT	53
REFERENCES	54
GENERAL REFERENCES ON THE MEASUREMENT OF RADIOACTIVITY	64
APPENDIX I. FORMULAS FOR GEOMETRY CALCULATIONS. . . .	67
APPENDIX II. CORRECTIONS FOR COUNTER NONLINEARITY . .	69

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INTRODUCTION

The general availability of radioisotopes has placed a powerful tool in the hands of investigators in many fields of science, and it is the purpose of this report to aid these workers in the design of meaningful experiments utilizing radioactivity. An understanding of the various factors that may contribute to errors in measurement is essential for the choice of appropriate techniques and measuring devices and for the proper evaluation of experimental data. This report is not intended as a treatise on the measurement of radioactivity, but it is hoped that the presentation of a handy guide to some of the problems involved and the techniques generally employed in avoiding or correcting for them, together with references to the pertinent literature, will provide an adequate orientation and introduction to the subject.

GENERAL CONSIDERATIONS

Although a variety of techniques is available for the assaying of the radioactive strength of samples of various kinds, the methods discussed herein are limited to those that employ particle counting and do not refer to photographic methods, calorimetry, charge collection, colloid deposition, crystal coloring, and other techniques of rather specialized application. Different techniques may, of course, involve somewhat different error analyses; however, the problems associated with the determination of the absolute disintegration rate of a radioactive source by counting techniques afford an excellent example of the commonly encountered difficulties. Such a determination involves a knowledge of the decay scheme of the radionuclide and the efficiency of the detector for the particles or rays emitted by the source. Thus it is necessary to know the fraction of the radiation leaving the source that enters the sensitive volume of the detector ("geometry factor") and the intrinsic efficiency of the detector for the radiation. The interactions of radiation with matter alter the simple geometrical relationship between the source and detector through processes of absorption and scattering in the material of the sample itself, its support, any covering material, the space between the sample and the counter, the counter window,

and the housing of the detector unit. These effects are complex and depend critically on the size and material of the sample and its support, and on the energy and type of the radiation. Special techniques are required to reduce these effects and limit errors in the determination of disintegration rates to the order of a few per cent.

For many purposes an experiment can be designed so that relative counting rates of samples of a particular nuclide will suffice. It is then only necessary to count under nearly identical conditions to obtain precisions of the order of one to two per cent. This criterion, in practice, is relatively simple to meet, and, whenever possible, comparative counting is therefore recommended. Of course, if it is necessary to compare the activities of sources that differ in chemical or physical nature or in the energy of their radiations, these differences must be taken into account.

Much time and effort may be saved by the judicious application of published correction factors for the individual and combined effects of backscatter, sample thicknesses, etc., on counting determinations. However, it is extremely important to be aware of the sensitivities of the effects to experimental conditions. When high accuracy is desired, it is usually advisable for each investigator to determine the correction for the conditions under which his experiments were carried out or to employ techniques that reduce the magnitudes of the effects materially.

In some cases, the radioactivity of the nuclide of interest may be complicated by the presence of isotopic or genetically related species that are also radioactive. Corrections for the presence of these species may be made, in general, by taking advantage of the differences in the half-lives, energies, and, perhaps, types of radiation involved. Even in the case of a pure nuclide, different types of radiation may be emitted (e.g., β , X, or γ rays) and their contributions to the observed counting rates must be evaluated.

Any detector will record counts, even in the absence of a sample, due to the presence of cosmic rays and of ionizing radiation in the materials of construction and in the laboratory air (thoron, radon, etc.). This background rate may vary with laboratory conditions and must be determined under conditions duplicating those under which the sample is counted. It must be subtracted from the sample counting rate. Since the background rate limits the sensitivity of the detector, it is advisable to reduce this rate to a level that is small in comparison with sample-counting rates. The background rate is dependent on the size of the detector. For standard end-window Geiger-Müller and proportional counters (about $1\frac{1}{4}$ in. in diameter by 2 in. long) backgrounds of 30 to 50 counts per minute are common. This rate may be reduced by a factor of two to three by the common practice of enclosing the detector in a lead or iron housing of one to two inches in thickness.

Further shielding of beta counters from penetrating cosmic radiation may be achieved by surrounding the detector with a ring of additional counters connected so that the detector will not record a count that is coincident with one in the protective ring. The entire assembly is enclosed in an iron, lead, or mercury shield. Such an arrangement - referred to as anticoincidence shielding⁽¹⁾ - may reduce the background rate to as low as 1 to 2 counts per minute. Additional corrections may be necessary for the variation in efficiency of the detector with time (as determined by the routine counting of a suitable standard) and for the finite resolving time of the detector (see Appendix II).

Radioactive decay is a random process, and inherent fluctuations in the disintegration rate of any source limit the ultimate accuracy of counting data. These fluctuations follow well-known statistical laws, and the statistical accuracy of a set of data can be readily calculated.

BETA COUNTING

End-window Counters

Although many specially designed counters are employed to satisfy particular needs,⁽¹¹⁸⁾ the end-window Geiger-Müller counter has probably been the most universally used detector for beta radiation. Consequently, much of the experimental work on the problems of beta counting has been carried out with this detector. Although the Geiger-Müller counter still finds useful application, it is generally being replaced with the flow-type, end-window proportional counter, which has many advantages. Since it is operated at atmospheric pressure, a thin window (usually 0.5 to 1.0 mg/cm² plastic foil* coated with a conducting film of Aquadag or evaporated metal) can be used; hence the range of application can be extended to betas of lower energy while the convenience of counting samples externally is retained. Geiger-Müller counters are usually operated at a pressure of ca 10 cm Hg, and window thicknesses greater than about 2 mg/cm² are most usual. Moreover, the proportional counter permits a much wider range of source strengths to be used, since the dead-time losses are of the order of 1% per 10⁵ c/m compared with losses of about 0.5% per 10³ c/m in a Geiger-Müller counter. Proportional counters require associated electronic equipment of somewhat greater complexity than Geiger-Müller counters (e.g., stable high-voltage supplies and nonoverloading amplifiers), but these are readily available. Considerable experience with proportional counters has proved them to have long lifetimes and excellent stability.

Fortunately, the size and shape, as well as the commonly employed holder and shelf arrangement, are very nearly identical for the Geiger-Müller and end-window proportional counters, and the extensive studies of various effects in beta counting carried out with Geiger-Müller counters are in most cases directly applicable to the proportional counter. Of course, there will be differences in some cases in which the window thickness plays a significant role, but even here the effects will be qualitatively the same. It cannot be overemphasized that the greatest contribution of the many studies of beta counting reported in the literature lies in their elucidation of the complex effects encountered and the presentation of methods of correction. The quantitative data presented are useful for approximate corrections and general orientation. However, for the highest accuracy, the quantitative factors should be determined for the particular experimental arrangement being used.

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* For example, rubber hydrochloride, available from Originality House, Inc., 480 Avalon Ave., Akron, Ohio, or Mylar (polyethylene terephthalate), available from E. I. du Pont de Nemours & Co., Wilmington, Delaware.

The importance of absorption and scattering phenomena in the detection of beta particles was pointed out as early as 1910 by Kovarik and others.⁽²⁾ Many aspects of the problem of the determination of absolute disintegration rates from counting data have since been investigated experimentally. The complexity of the problem has prevented successful theoretical development, and the magnitudes of the effects must usually be determined from empirical data. Although it is not always necessary to obtain absolute disintegration rates, the investigator should be aware of the factors that may contribute to errors in counting determinations.

When particle emission accompanies each decay, a counting yield, $\underline{Y}$, may be defined⁽³⁾ as the ratio of the net counting rate observed (corrected for resolution losses and background activity) and the true disintegration rate. Thus,

$$\underline{Y} = \frac{\text{counts per minute (c/m)}}{\text{disintegrations per minute (d/m)}}$$

For simplicity, the present discussion is limited to the case of a simple beta emitter (such as P^{32} or C^{14}). More complex decay schemes with γ rays, X rays and conversion electrons, as well as beta branching, will be discussed later. Although the factors that enter into the determination of $\underline{Y}$ are not simple and not easily separable experimentally, they can, in principle, be treated independently. Then,^(3,4)

$$\underline{Y} = \frac{c/m}{d/m} = (I) (G) f_W f_A f_C f_B f_H f_S \quad ,$$

where I = detection probability, G = geometry factor, f_W = factor for the absorption effect of the counter window and air between source and window, f_A = factor for the scattering effect of the air, f_C = factor for the effect of any covering material over the sample, f_B = factor for the effect of backscattering by the material supporting the source, f_H = factor for the effect of the sample and tube-supporting structure and the walls of the housing or shield in scattering counts into the counter, and f_S = factor for the effect of the finite mass of the source ("self-absorption" and "self-scattering").

I (detection probability). Ideally, every particle that passes through the window of an end-window Geiger or proportional counter will form at least one ion pair, initiate a Townsend avalanche, and register a count. Actually, however, there is a finite probability that an ion pair will not be formed. This probability may be calculated⁽⁴⁾ and is dependent on the specific primary ionization constant of the counter gas, the pressure of the gas, the energy, and the path length of the particle through the volume of the counter. In general, the detection probability is close to unity, and no corrections are required except for work of extremely high accuracy.

For cases of high geometry and energetic betas, the path length obliquely through the corners of the counter may be too short to produce sufficient ionization to register a pulse. This will result in a shift of the knee of the voltage plateau to higher values. Defining apertures in front of the window may counteract this effect by collimating the particles, but these apertures may also introduce scattering effects from the edges. It may be more advisable to reduce the geometry of counting by increasing the source to tube distance if the added path length does not introduce an absorption problem.

Effects such as these illustrate the care that should be exercised in the choice of operating voltage. Voltages just above the plateau knee for weak betas may be below the plateau for energetic betas, since the specific ionization of electrons decreases with increasing energy over the energy range generally encountered. On the other hand, the resolving time of the counter increases with increasing voltage along the plateau, and a voltage that ensures a high detection probability may introduce higher resolution losses.

In practice, this problem is generally met by determining the geometry of the source position with standards having approximately the same energy and counting rate as the sources to be measured. In this case, a "geometric efficiency" is actually determined, in which the detection probability and resolution losses cancel.

G (geometry). Radiation is emitted isotropically from a point source and, in the absence of absorption and scattering phenomena, the fraction of the solid angle of 4π radians subtended by the sensitive volume of the counter at the source will represent the fraction of the emitted radiation that is counted. This fraction is known as the "geometry" or "physical geometry" and is determined simply by the dimensions of the system. Formulas may also be derived for the geometry of extended sources. These are given in Appendix I.

The geometry is generally determined by counting standards of known disintegration rates (determined by coincidence counting, 4π counting, or other techniques) in the desired position. In order to minimize the corrections for absorption and scattering, standards are prepared in thin, uniform deposits on thin backings.^(4,5,6,59e) The ratio of the counting rate of the standard (suitably corrected for absorption and scattering effects, as well as for background activity and any resolution losses) and the known disintegration rate gives the geometric efficiency.

It has been reported^(4,5,7) that the observed geometric efficiency is slightly lower than that calculated from the dimensions of the counting system. This may arise from the collection of part of the charge on the ball at the end of the wire, but it has also been interpreted as indicating that the

active volume of the counter begins a few millimeters behind the window. A "defined geometry" technique employing a brass collimator has been suggested(7,8,9) to avoid this difficulty. However, Fried et al.,⁽¹⁰⁾ observed significant increases in the curvature of absorption curves of Co^{60} and RaE at low absorbed thicknesses and demonstrated that, when appropriate corrections for absorption in the window and gas path are applied, the observed and calculated geometries do, indeed, agree within the accuracy of the measurements.

The geometric efficiency for extended sources has been evaluated empirically and shown to agree well with the calculated geometry.^(4,11-14)

Although, as previously pointed out, it is desirable to use standards having the same energy as the sources of interest, errors in the extrapolation of absorption curves (see section of f_W) to zero thickness may result from differences in the shapes of the beta spectra of two nuclides with the same maximum beta energy. Manov⁽¹⁵⁾ suggests that many standards may be required for the accurate calibration of sources of general interest and has reviewed the status of the work on the standardization of radioactive sources to 1954.

f_W (factor for air and counter window absorption). Since beta particles are easily scattered in passing through matter, an absorbing material placed between the source and detector may reduce the intensity of radiation reaching the counter by true absorption or by scattering particles out of their normal paths toward the detector. On the other hand, some particles may be scattered into the detector, increasing the apparent intensity. These effects are critically dependent on the relative positions of source, absorber, and detector.^(4,16-19) If the absorbing material is close to the counter window, scattering effects are reduced, and true absorption effects are observed. The fractional reduction in counting rate due to the window and air, f_W , will be dependent on the energy of the radiation and the thickness* of the window and air path. The factor f_W is determined from an absorption curve by extrapolation to a negative value of added absorber corresponding to the average thickness of air and window (i.e., to zero total absorber) (see Figure 1). In determining such a curve, the absorbing foils should always be placed close to the counter window. Since the average atomic number of air and plastic counter window materials is close to that of the conveniently prepared aluminum absorbers, aluminum is usually used for absorbing foils. The fact that the air is an extended absorber and scattering medium is neglected here, but it is taken into account in the discussion of the factor f_A .

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*It is convenient to express the "thickness" of absorbing material in terms of surface density (mass per unit area), since on this basis the absorption of beta particles is nearly independent of the nature of the material, particularly for absorbers of low atomic number.

Figure 1

Illustration of extrapolation of initial portion of a typical absorption curve to correct for 5.0-mg/cm² absorption in tube window and air path.

Serious errors may arise in the extrapolation of absorption curves to zero total absorber. It is often stated that the absorption of beta particles is nearly exponential. This statement can be misleading. The complex nature of beta spectra and absorption and scattering phenomena do, in many cases, give rise to absorption curves that are exponential over the initial portions. In fact, initial half-thickness values or absorption coefficients are conveniently used to determine corrections for absorption analytically (i.e., $N = N_0 e^{-\mu t}$). (For approximate calculations, a curve of initial half-thickness versus energy may be useful and is given in Figure 2.) Careful determinations of absorption curves, however, particularly when using the thin-window proportional counters, have indicated that the exponential character of beta absorption is not a valid generalization, and the absorption curves of many nuclides indicate initial curvatures. Zumwalt⁽⁴⁾ has suggested a formula for calculating the extrapolated counting rate in terms of the initial slope of the absorption curve and the curvature. In determining the factor f_W graphically, it is obviously necessary to obtain a sufficient number of points on an absorption curve as close to zero absorber as possible in order to increase the accuracy of the extrapolation. Since the absorption is nearly exponential in many cases, it is convenient to use semilog paper for the graphical presentation.

f_A (factor for scattering effect of air). The effect of the air in scattering particles into the counter was determined by Zumwalt⁽⁴⁾ indirectly by the use of thin polystyrene films placed between the source and counter. The magnitude of the effect increases with distance from the counter and with decreasing energy of the beta particle. With the sample on the first shelf position (0.5-cm air path), he found that the factor varied from 1.001 for UX_2 (2.3-Mev β) to 1.005 for Co^{60} (0.3-Mev β), and on the fourth shelf (5.3-cm air path), from 1.012 for UX_2 to 1.060 for Co^{60} .

Figure 2

Energy versus half-thickness in aluminum for β particles. [These data were obtained with samples weighing about 20 mg and mounted over a 2-cm² area on filter paper and covered with 3-mg/cm² cellophane. They were counted on the first shelf (geometry factor = 25% of 4π) of a standard Geiger counter (tube window = 3.0 mg/cm²).]

f_C (factor for effect of sample covering). In many cases samples are covered with thin, protective plastic films. Since these are close to the sample, scattering effects are important, and the counting rate is usually increased somewhat over what would be observed if only absorption were taking place. This "forescattering" effect was first observed and qualitatively explained by Kovarik⁽²⁾ (also see references 4 and 20). The effect is a function of the thickness and atomic number of the absorbing foil, the energy of the beta radiation, and the distance between the sample and the detector. For a given covering material (3 mg/cm² of polystyrene), Pappas⁽²⁰⁾ found that the forescattering factor was negligible for a source-to-counter distance of 5.3 mm and equal at 21.3, 37.3, and 53.3 mm. He defines the factor as the ratio of counts with the cover on the sample to that with the cover near the counter window (where it acts only as an absorber). The factor decreases with increasing energy of the beta particles from about 1.11 at 0.2 Mev to ~ 1.01 at 2.2 Mev. Zumwalt⁽⁴⁾ examined the forescattering effect (defined as the difference between the counting rate with absorber directly on the source and near the window expressed as a percentage of the counting rate of the sample with no absorber) for second and fourth shelf positions with various beta rays and absorbers. In general, the forescattering effect increases to a maximum with increasing thickness of absorber and then decreases again. The maximum tends to shift toward thicker absorbers as the beta energy increases and as the atomic number of the absorber decreases. The initial slope of the forescattering curves increases with decreasing beta energy and with increasing atomic number of the absorber. The maximum forescattering increases somewhat with decreasing geometry.

Studies of the angular distribution of scattered radiation⁽²¹⁾ indicate that the spatial distribution follows a cosine law. The isotropic emission expected from an isolated source is modified so that the radiation is concentrated in the direction of the detector.

Corrections for the effect of any covering on the sample may be made for the first shelf position by considering the cover as added absorber and extrapolating the absorption curve as in the case of the air and counter window correction (see f_W). At lower geometries, the scattering effects must also be taken into account. These act to compensate the loss by absorption in the cover and, in many cases, the two effects nearly cancel, giving a value of f_C near unity.

f_B (factor for effect of backscattering). Many experimental studies of the backscattering of electrons have been reported.^(2,4,5,22-32) The data obtained indicate the dependence of the effect on the nature and thickness of the scattering material, the electron energy, and the geometry of the source.

Figure 3

Typical backscattering
"growth" curve.

The counting rate of a sample is increased by contributions from radiation initially directed away from the counter but scattered back by the material on which the source is mounted. This contribution increases with the thickness of the backing to a saturation value that occurs at a thickness equivalent to about 15-25% of the range of the beta particles in the material (see Figure 3). (The fraction of the range at which saturation backscattering occurs decreases with increasing beta energy and, to a lesser extent, with increasing atomic number of the scatterer.) The per cent increase in counting rate at saturation increases with increasing atomic number of the scatterer (Figure 4). When appropriate corrections are made for the difference in absorption of the backscattered radiation (which is degraded in energy) in the air and counter window, it is seen that the saturation backscattering for a given scatterer is independent of energy over the range investigated (0.17-1.7 Mev).^(4,5,26)

Most data on backscattering have been obtained with sources on thin films and scattering materials placed very close to or in contact with the film. Significant increases in the backscattering effect are obtained with the sources placed directly on the scattering materials, possibly as a result of surface imperfections.

The degradation in energy that takes place as a result of inelastic collisions in the process of backscattering is a function of the atomic number of the scatterer.^(4,17,26,30)

Figure 4

Saturation backscattering of β radiation as a function of the atomic number of the backscattering material. The solid line represents data for the source directly on the surface of the backscatterer, and the broken line represents data for the source on a thin film separating it from the backscatter.

Experimental data on the energy degradation of backscattered radiation indicate somewhat greater energy degradation on scattering from materials of low atomic number than from high. These data are presented in Figure 5 in terms of the relative half-thicknesses of scattered and unscattered radiation.

Figure 5

Energy degradation of backscattered radiation as a function of the atomic number of the backscattering material.

Experimental investigations of the angular distribution of backscattered radiation have shown it to be anisotropic. (4,21,25,27,32) The angular anisotropy is a function of the atomic number of the backscatterer, (25,27,32) and Seliger (27) has shown that the distribution can be conveniently interpreted on the basis of "diffusion" and "side-scattering" effects, the former increasing with atomic number Z , whereas the latter is relatively constant. For material of low Z the energy degradation is much larger at a scattering angle of 90° than at 10° , whereas for high Z both are about the same.

f_H (factor for the effect of the housing). Scattering from the walls of the counter shielding and sample support structure contributes negligibly to the observed counting rate of a sample mounted on thin backing except for positions near the base of the housing.⁽⁴⁾ The effect increases with energy of the beta ray, and on the fourth shelf (11 mm from the base of an aluminum-lined lead shield and about 53 mm from the tube window) Zumwalt observed an increase from a factor of 1.045 for Co^{60} to 1.102 for UX_2 .

This effect is dependent on the materials used for construction of shield linings and sample supports. Materials of low Z should be employed whenever possible in order to reduce the scattering.

f_S (self-absorption and self-scattering factor). It is not always convenient or possible to prepare samples that are effectively "weightless." In many experiments, samples of low specific activity are encountered or carriers are used, and it is necessary to count samples of appreciable weight. In such samples, scattering and absorption of radiation takes place within the source material itself. The effects depend in a complex manner on sample thickness, the effective atomic number of the source material, the energy of the radiation, and the geometry. The largest error in the determination of beta disintegration rates probably lies in the determination of f_S .

Although a number of attempts has been made to generalize the effects of self-absorption and self-scattering, and some equations and graphs have been published,^(25,33-55,126) the data are, in general, applicable only to experimental conditions duplicating those under which the effects were studied. The data are, of course, extremely useful in illustrating the nature of the effects and can be used for approximate corrections. For more accurate work, however, the corrections should be determined empirically by each investigator.

Self-absorption in thick samples is fairly well represented by an equation that assumes exponential absorption and no self-scattering. The activity A_t observed from a sample of thickness t (in mg/cm^2) is given by^(39,40)

$$A_t = A_0 \frac{(1 - e^{-\mu t})}{\mu t} ,$$

where A_0 is the activity that would be observed from a weightless source, and μ is the mass absorption coefficient measured in cm^2/mg . For geometries of less than 2π , the angular distribution of scattered radiation gives rise to the "forescattering effect" discussed in connection with added absorbers. With increasing weight of a uniformly radioactive source, the specific activity (counts/minute/mg) rises initially as a result of self-scattering, reaches a maximum (at about 5-10% of the range of the β radiation), and then decreases in accordance with the self-absorption equation (Figure 6).

Figure 6

Illustration of self-absorption and self-scattering effects as a function of sample weight.

It is obvious that if sufficient data are not obtained at small thicknesses, or if the simple self-absorption equation is applied to data beyond the maximum, the extrapolated counting rate at zero weight may be in error. In an extreme case the error may be as much as a factor of two.

Baker and Katz⁽⁵²⁾ have shown that such a curve can be described as the product of self-scattering and self-absorption curves. The self-scattering curve is determined from measurements of the angular distribution of the radiation from thick sources and is expressed in terms of the ratio $N_{0,x}/\bar{N}_x$, where $N_{0,x}$ is the activity measured in a direction normal to the plane of the sample of thickness x and $\bar{N}_x$ is the activity averaged over all angles. Although sufficient data to establish a dependence on atomic number were not obtained, a single curve described $N_{0,x}/\bar{N}_x$ as a function of sample thickness (expressed as fraction of the range). Walton⁽⁵³⁾ derived equations describing the combined effects of self-absorption, self-scattering, and back-scattering, which account fairly well for observed results. It is significant to note that sources with smooth flat surfaces were essential in order to obtain reproducible results in Walton's work. This illustrates the necessity of maintaining standardized conditions in relative determinations of counting rate. A similar study has been carried out by Lerch.⁽⁵⁴⁾

The foregoing discussion of the factors that may contribute to errors in beta counting illustrates the complexity of the problem of determining the disintegration rate of even a simple beta emitter from a thick source. When other radiations from the nuclide of interest or genetically related species are present, their contribution to the observed counting rate must be evaluated. In general, absorption techniques are combined with decay measurements to analyze multicomponent systems, and the radiations of interest are corrected for the effect involved. These measurements may introduce additional errors arising from uncertainties in the half-lives of the activities involved and difficulties in analyzing absorption curves (Figure 7).

Figure 7
Analysis of a complex
absorption curve.

Suggested Techniques for End-window Counting

Absolute Beta Counting. The most accurate method of determining beta disintegration rates, by means of end-window counters, is to prepare essentially weightless sources mounted on thin backing and to count in a position of known or calibrated geometry. If the geometry has been determined by the use of some standard (such as RaE⁽⁶⁾) or is calculated, it will be necessary to apply corrections for absorption and scattering effects. Of course, if a geometric efficiency for the radiation has been determined, no corrections will be needed (other than background and any resolution loss). Such techniques should give errors of the order of 5% or less.

When it is necessary to determine absolute disintegration rates from thick sources within a reasonable degree of error, e.g., one of the order of 10% or less, it is essential to prepare calibration standards. A high specific activity source of the activity of interest should first be prepared and its disintegration rate determined by 4π counting, coincidence counting (if applicable), or the counting of a weightless source on thin backing in a position of known geometry. Equal aliquots of the activity are added to solutions containing varying known amounts of the element of interest. The compound to be measured is precipitated, the chemical yield determined, and the sample mounted and counted under the desired conditions. The observed counting rates are corrected for chemical yield, background, and resolution losses. A comparison of the observed activity of these samples with the known disintegration rate of the aliquot added gives an overall counting efficiency for the conditions of the measurement.

The counting efficiency for each sample may be plotted graphically as a function of sample weight, and a calibration curve that can be readily applied to the determination of disintegration rates of other samples measured under the same conditions will thereby be obtained. It is, of course, important to ensure reproducibility of the sample-mounting technique by determining a number of samples at each weight. Similar curves must be obtained for each activity or compound to be measured.

If it is not possible to obtain a source of high specific activity, or if errors of the order of 20% will suffice, correction factors or equations reported in the literature^(25,31-55,126) may be used. In general, these factors have been determined under conditions that should not be difficult to reproduce rather closely.

Examples of such correction factors for S^{35} ($\beta^- = 0.167$ Mev, precipitated as $BaSO_4$) and P^{32} ($\beta^- = 1.7$ Mev, precipitated as $MgNH_4PO_4 \cdot 6H_2O$) are given in Figure 8. The curves represent data⁽¹¹⁹⁾ for precipitates of different weights collected over a 2-cm² area on filter paper, mounted on cardboard cards, and covered with rubber hydrochloride 0.5 mg/cm² thick. The samples were counted in the first shelf position (geometry factor = 36% of 4π) of a standard end-window flow proportional counter assembly using a counting tube with a 0.6-mg/cm² window. The disintegration rate of an aliquot of a solution of the radionuclide was determined by 4π counting. Equal aliquots were incorporated in precipitates of varying weight and the counting rates were determined. The observed counting rates were corrected for the chemical yield in the precipitation (i.e., the fraction of the added carrier actually mounted) and for the absorption effects of the sample covering, the air path, and the counter window. The correction factor F represents the ratio of the disintegration rate to the corrected counting rate of the samples.

Figure 8

Empirical correction factors F for S^{35} and P^{32} to obtain disintegration rates from observed counting rates.

Figure 9 gives the correction factor F as a function of β -particle energy for precipitates of 10-mg/cm² thickness. Differences in the average effective atomic number of precipitates other than those used in determining

the curve of Figure 8 may be ignored for approximate calculations when errors of the order of 10% may be tolerated. The factor is essentially constant for source thicknesses over the range from 5 to 15 mg/cm².

Figure 9

Empirical self-absorption, self-scattering correction factor as a function of beta energy for 10-mg/cm² sources.

Bayhurst and Prestwood⁽¹³⁷⁾ have pointed out that the average beta energy, which is dependent on the shape of the beta spectrum, is a better parameter for correlation with counting efficiency than the maximum energy as used in Figure 9. They have prepared a number of standardized sources and constructed counting-efficiency curves as a function of average energy and sample weight by a procedure similar to that outlined above. They estimate an error of the order of 3% for the determination of absolute disintegration rates with this technique.

In some instances, if the activity level permits, a solution of a thick sample may be dissolved and relatively thin sources prepared from small aliquots.

Relative Counting. The most important consideration in relative counting is the reproducibility of sample preparation and mounting techniques. In particular, sample areas and thickness should be the same. Reproducibilities with about 2% error may be expected for the relative counting rates of properly prepared samples. A number of techniques for preparing reproducible samples has been reported.^(38,44,45,47,48,51,53,120,121,126) In comparing the counting rates of samples which may differ in chemical composition, weight, size, mounting, etc., corrections must be applied as previously outlined and consequently the precision of the result may be somewhat less. In general, it is desirable to prepare either very thin samples, in which the corrections are minimized, or samples thick enough to be approaching saturation of the effects, since such samples can be most readily made reproducibly.

2 π Counting

Counting under conditions of a solid angle of 2 π radians may be carried out with external or internal samples. Since the effects of the anisotropic distribution of scattered radiation are considerably reduced

in magnitude at such high geometry, the major effects of concern are true self-absorption and back-scattering (including that from the sample itself). These are treated in the same manner described for end-window counting, namely, by reducing the effects, if possible, or by establishing empirical correction factors.^(55-57,125)

For samples of low activity, 2π counting is particularly advantageous and numerous instruments have been utilized for this purpose. Samples may be mounted externally by wrapping around glass-walled cylindrical Geiger-Müller counters,⁽¹³⁴⁾ or internally in a flow-type proportional counter or screen-wall counter.⁽⁵⁸⁾ The internal counters are, of course, desirable and sometimes necessary for the counting of very low-energy betas. These instruments may be calibrated as described in the previous section to determine a counting yield (or efficiency).

A typical efficiency curve⁽¹¹⁹⁾ for a flow-type internal proportional counter using weightless samples mounted on 5-mil Pt foils is shown in Figure 10. The fact that the efficiency falls below 50% at very low energies is probably the result of self-absorption in the sources. It is extremely difficult to prepare sources that are essentially "weightless" for radiation of such low energy. Somewhat different results would be obtained with different mounts as a result of backscattering differences. Backscattering corrections have been determined⁽¹²²⁻¹²⁴⁾ for internal (2π) flow proportional counters as a function of the atomic number of the mounting plate; such a curve⁽¹²²⁾ is given in Figure 11, where the backscattering factor F_B represents the ratio of the observed counting rate of a known aliquot of a solution of the radionuclide to one-half of the known disintegration rate of the aliquot (determined by 4π counting). In many designs of 2π counters, materials of construction are close to or in contact with the sample mount. Thus, it may not be possible to eliminate or minimize backscattering by the use of thin films for mounting samples, and mounting plates thick enough to give saturation backscattering should be used. Even with the same mount material, some differences in backscattering may be observed because of the differences in surface conditions, and where high precision is desired this should be borne in mind.

Figure 10

Counting efficiency of an internal (2π) flow proportional counter for samples mounted on 5-mil Pt.

Absolute beta-disintegration rates may be determined by using internal (2π) counting technique, with suitable calibration, to an accuracy involving errors of the order of a few per cent.

Figure 11
Backscattering at 2π Geometry

Suttle and Libby⁽⁵⁶⁾ have shown that, under conditions of close cylindrical geometry, such as obtains in the use of a screen-wall counter, the assumption of purely exponential absorption is valid for thick sources of simple beta emitters. For a sample thick with respect to λ , the reciprocal of the mass absorption coefficient of the radiation, the absolute specific radioactivity in disintegrations per minute per gram of sample is given by

$$\sigma = RG/A\lambda \quad ,$$

where R is the observed counting rate in counts per minute, A is the area of the sample in square centimeters, λ is the reciprocal of the absorption coefficient of the radiation in units of grams per square centimeters, and G is the ratio of 4π to the solid angle subtended by the sensitive counter volume at the source. (It is assumed that G is not seriously dependent on the depth of sample and has been averaged over its area.) With this technique, absolute disintegration rates may be obtained with an error of about 5%.

4 π Counting

If one event were recorded for each disintegration of a radionuclide, a direct measure of the absolute disintegration rate would be readily obtained. In practice, the 4π counter comes closest to achieving this ideal condition. Designs of 4π counters, utilizing twin, facing Geiger, or proportional counters, with the sample mounted between them, and multiple counter arrangements,⁽⁵²⁾ have been described.⁽⁵⁹⁾ Methods based on charge collection have also been reported.⁽⁶⁰⁾ The sample may also be homogeneously distributed through the detector in the gas phase or in solution (see sections on Internal Gas Counting and Beta Scintillation Counting).

When the two halves of a 4π counter are connected in parallel, coincident events [such as betas, conversion electrons, secondary (delta) rays, X rays, or gammas] will be recorded as a single pulse. The detection efficiency of Geiger and proportional counters for particles is very close to 100%, so that if particle emission (which may be β^- , β^+ , conversion, or Auger electrons) accompanies every disintegration (or a known

fraction of the disintegrations), the 4π counter, in principle, will record the disintegration rate directly, regardless of the complexity of the decay scheme. If excited states occur with lifetimes longer than the resolving time of the instrument, these will, of course, be recorded as independent events, and their contribution to the observed count must be determined from a knowledge of the decay scheme. Likewise, the presence of isotopic or genetically related radioactivities of the nuclide of interest will contribute to the observed counting rate, and their contribution must be evaluated. As is necessary in all counting methods, background activity and resolution losses must be corrected for.

Although the 4π counter offers a versatile and convenient method of determining absolute disintegration rates, a few sources of error must be considered, namely, absorption of radiation in the mounting film that separates the two counters, the possibility of a counting efficiency of less than 100%, and self-absorption in the sample.

Pate and Yaffe⁽⁵⁹⁾ have carried out a systematic survey of 4π counting techniques with twin, facing proportional counters. They have examined the problem of source-mount absorption correction and have pointed out errors in previous publications in connection with this correction. Three techniques have been used to correct for this effect: (1) a "sandwich" procedure,⁽⁶¹⁾ in which the counting rate is determined for the sample on a mount of known thickness and another determination is made with a film identical to the mount covering the sample; (2) a calculation consisting of simplified assumptions of scattering and absorption effects and measurements of 2π and 4π single and sandwiched-film counting rates⁽⁶²⁾; and (3) an absorption curve method⁽⁶³⁾ in which the source is mounted on a thin film (5 to 10 $\mu\text{g}/\text{cm}^2$) and the counting rate is observed as a function of increasing mounting-film thickness. Increased thicknesses of film are obtained by allowing thin films of known thickness to adhere to the back of the mounting film. The "absorption curve" is extrapolated to zero thickness to obtain the disintegration rate.

For beta energies above a few hundred kev, any of these methods work quite well, since the corrections are small. However, at lower energies the absorption curve technique appears to be the most accurate. Pate and Yaffe estimate that disintegration rates can be determined within an error of about $\pm 0.2\%$ for energies as low as 67 kev and probably with higher accuracy at somewhat higher energies. Their data are reproduced in Figure 12 in which the ratio of observed counting rate to disintegration rate (N_T/N_O) is given as a function of beta end-point energy for a series of film thickness. These data may be used directly to obtain the source-mount correction for beta rays of known energy. It should be remembered that beta-ray spectra may differ in shape even for identical end-point energies, and, unless the correction is to be applied to a nuclide for which data is given in Figure 12, the accuracy of the disintegration rate value will not be so high as if an empirical determination of the absorption curve were made.

Figure 12

Absorption parameter (N_T/N_0) as a function of beta end-point energy for a series of film thicknesses.

Geiger-Müller and proportional counters have sensitive volumes that are very nearly 100% efficient, but, as previously discussed, there may be regions of low field strength and a consequent loss in detection efficiency. This is particularly true in internal counters, in which the source is mounted on a nonconducting film on which charge may accumulate and cause field distortion. Thin evaporated metal coatings should be used on all mounting films.^(59b)

The most difficult correction to make is that for self-absorption in the source. The problem of preparation of very thin sources has plagued many fields of investigation, particularly when known aliquots must be deposited. Examinations with optical and electron microscopes show evaporated sources to consist of small clusters of crystals. Additional solids (as carrier) might fill in the spaces between clusters to give a more uniform, but not necessarily thicker, source. Thus the technique of determining a self-absorption curve by depositing equal aliquots containing increasing weights of carrier may not be applicable. Optical measurements of S^{35} and Co^{60} sources^(59e) indicate that with increasing weight of carrier the number and size of crystals increase. The problem of source preparation is not easily solved, and, at present, the best that can be done, if a self-absorption curve does not appear applicable, is to examine sources of low-energy betas and estimate the self-absorption loss from the density of the deposit. Corrections of the order of a few per cent may be expected, e.g., for Co^{60} (0.3 Mev). They may, of course, be less for more energetic and greater for less energetic radiation.

Methods for preparation of thin films and samples are given by Slätis,⁽¹³⁵⁾ Povelites⁽¹³⁸⁾ and Parker,⁽¹³⁹⁾ et al. The most satisfactory method for preparation of thin uniform samples appears to be by volatilization from a hot filament.^(59e, 64, 140) An electrostatic spray technique⁽¹⁴¹⁾ and the use of micro-exchange columns⁽¹⁴²⁾ have proved useful for a number of nuclides.

The 4π counting technique is the most generally applicable method for the determination of disintegration rates of radioactive sources and is recommended for the most precise work. Methods of measurement for the calibration of a number of radioactive sources prepared at the National Bureau of Standards are reviewed by Seliger and Schwebel.⁽⁶⁵⁾ This report illustrates the application of the 4π , coincidence, and internal gas-counting techniques (see below).

Internal Gas Counting

Another type of 4π counting may be achieved by incorporating the activity in the gas phase with the counting mixture in a Geiger or proportional counter⁽⁶⁶⁻⁶⁸⁾ or by the use of ionization chambers.⁽⁶⁹⁾ For the determination of absolute disintegration rates, cylindrical tubes are filled with measured quantities of counting and radioactive gases. Each measurement requires a new filling, and the counter characteristics must be checked to ensure proper operating conditions for each determination.

In addition to the corrections for background and resolution losses, the effects of field inhomogeneity at the insulated ends of the sensitive volume (end effect) and the escape of radiation originating near the cathode wall before causing ionization (wall effect) must be considered. End-effect corrections for brass-wall counters having flat Lucite ends have been investigated as a function of the ratio of length to diameter of the counter.⁽⁷⁰⁾

The end effect is dependent on the tube construction and may be determined from measurements in two counters of identical construction and diameter but different length. The wall loss is a function of the energy of the radiation and is inversely proportional to the pressure in the counter and to the diameter of the tube. Backscattering and secondary electron emission from the wall tend to compensate the wall loss. The end and wall effects may be represented as additive correction terms on the actual bare wire length ($\underline{L}$) and tube diameter ($\underline{D}$). The active volume of the counter $\underline{V}$ is then given by

$$V = \pi(\underline{D} + \underline{d})^2 (\underline{L} + \underline{\ell})/4$$

where $\underline{d}$ and $\underline{\ell}$ are the corrections to the diameter and wire lengths, respectively. These corrections may be determined by measurements with counters of constant length and varying diameters, and vice versa, or by measurements with standardized gas samples.

The end effect may be eliminated by the use of paired differential counters,⁽⁷¹⁾ differing only in length, or by employing a movable and a fixed glass shield around the central wire anode.⁽⁷²⁾

The wall effect may also be determined by varying the pressure P and plotting the counting rate as a function of $1/P$. The curve may then be extrapolated to $1/P = 0$.

When proper care is exercised in internal gas counting, absolute disintegration rates may be determined with errors of the order of 1% and reproducibilities to within 0.5%.

External Gas Counting

Gas samples may be prepared in cells that can be counted externally in the same manner as a solid sample.^(73,74) Corrections for absorption and scattering effects as well as geometry must be made to determine disintegration rates. It is recommended that such cells be calibrated with gases of known disintegration rate if they are to be used for absolute measurements. For relative measurements, external gas counting may prove more convenient than internal counting, and it is necessary only to maintain reproducible conditions to obtain results of high precision.

Coincidence Counting

The coincidence counting technique^(9,75-77,136) may be used for absolute disintegration rate determinations in those cases in which beta and gamma rays are emitted in sequence within the resolving time of the apparatus (usually of the order of 10^{-6} second). Although the method may, in principle, be applied to any known decay scheme, it is generally limited to simple systems of a single beta particle followed by one or more gamma rays. The convenience and accuracy of the method rapidly decrease with increasing complexity of the decay scheme.

If a point source of activity (which decays through a single beta followed by a single unconverted gamma) is placed between a beta counter and a gamma counter (shielded so as to be insensitive to beta particles) and the counters are connected through a coincidence circuit, data sufficient for a determination of the absolute disintegration rate may be obtained. A schematic diagram of the apparatus is given in Figure 13. The counting rate in the beta counter, N_β , is given by

$$N_\beta = N\epsilon_\beta \quad ,$$

where N is the disintegration rate and ϵ_β is the overall efficiency of the counter for the beta particles (including geometry). Likewise,

$$N_\gamma = N\epsilon_\gamma \quad ,$$

where N_γ is the gamma counting rate and ϵ_γ is the efficiency of gamma counting. Since the coincidence counter registers only when a beta and gamma count occur simultaneously in the two counters, the counting rate of true coincidences, N_c , will be

$$N_c = N\epsilon_\beta \epsilon_\gamma$$

(assuming the coincidence circuitry has been adjusted to give unit coincidence efficiency). Then,

$$N = N_{\beta}N_{\gamma}/N_c$$

Figure 13

Schematic diagram of coincidence counting equipment.

The counting rates N_{β} , N_{γ} , and N_c must be derived from the observed counting rates by the application of a number of corrections. These include corrections for chance (random) coincidences, backgrounds, presence of conversion electrons, any angular correlation, and dead-time losses. In addition, for the coincidence method to be valid, at least one counter must have a constant efficiency (i.e., be equally sensitive) to radiations from all parts of the source. In general, the corrections are dependent on the particular detectors and electronic circuitry employed. It is advisable to use detectors that exhibit high efficiency for one radiation and low efficiency for the other. Thus, the beta counter should be a Geiger, proportional, or thin crystal scintillation counter, all of which are highly efficient for betas but have low gamma-counting efficiency. A number of types of gamma counter with fairly high efficiencies has been developed, among which the gamma scintillation counter using thallium-activated NaI is perhaps the best. Beta detection in the gamma counter is prevented by the interposition of a suitable absorber.

Since the pulses arriving at the coincidence circuit have finite widths, two independent counts (not associated with the same disintegration) may overlap and give rise to a coincidence pulse. This random coincidence rate is given by

$$C_R = 2\tau N_{\beta}N_{\gamma} \quad ,$$

where 2τ is the resolving time of the system, and N_{β} and N_{γ} are the observed singles counting rates in the beta and gamma counters, respectively. [It should be noted that, when the observed coincidence rate N_c is not small with respect to the observed singles rates N_{β} and N_{γ} , the foregoing expression for C_R must be modified to $C_R = 2\tau(N_{\beta} - N_c)(N_{\gamma} - N_c)$.]⁽⁷⁸⁾ The resolving time may be determined by the use of two independent sources suitably shielded from each other so that no scattered or coincident

radiation is registered. Any coincidences recorded, then, will be due primarily to a background of cosmic ray showers and the finite resolving time of the circuit. Sources of different strength should be used to determine any dependence of resolving time on counting rate. The coincidence rate is given by

$$M = B_C + 2\tau N_1 N_2 \quad ,$$

where B_C is the independently measured cosmic-ray background coincidence rate, and N_1 and N_2 are the singles counting rates.

Since it is desirable to keep the chance rate below the true coincidence rate, a limitation is imposed on the source strength. For C_R to be less than N_C , it is necessary that $2\tau N$ be less than unity.

Background activity must be subtracted from the observed counting rates. In the gamma counter, the background is determined simply from a measurement using a blank sample mounting. (For sources of high beta activity, bremsstrahlung in the beta absorber may also contribute to the gamma background.) In addition to registering normal background activity, the beta counter also records some gamma counts from the source. (In a more complex decay scheme, these gamma counts may give rise to γ - γ coincidences.) The background for the beta (and coincidence) channel is determined with an absorber (usually aluminum) shielding the beta counter so as to stop all betas from the source. The effect of the absorber on the gamma-counting efficiency in the beta counter must be determined empirically. With beta counters whose gamma-counting efficiency is low, the correction will be small.

Internal conversion of the gamma ray may take place. For the simple decay scheme under consideration (one beta followed by a single gamma), such internal conversion will contribute electron counts to the beta counter and N_β will be given by

$$N_\beta = N[\epsilon_\beta + a\epsilon_e(1 - \epsilon_\beta)] \quad ,$$

where a is the fraction of total disintegrations leading to conversion electrons (i.e., $a = N_e/N_\beta$) and is related to the internal conversion coefficient α by $a = \alpha/(1 + \alpha)$, and ϵ_e is the efficiency of the beta counter for the counting of conversion electrons. The last term, giving $\beta - e^-$ coincidences, can often be neglected if ϵ_β and ϵ_e are not too high. Likewise,

$$N_\gamma = N(1 - a)\epsilon_\gamma$$

and

$$N_C = N(1 - a)\epsilon_\gamma\epsilon_\beta$$

The foregoing equation has no term in ϵ_e since there can be no coincidences between the conversion electron and the converted gamma. Then,

$$\frac{N_\beta N_\gamma}{N_c} = N \left(1 + \frac{a\epsilon_e}{\epsilon_\beta} - a\epsilon_e \right) ,$$

where it is apparent that it may be advantageous to operate with $\epsilon_\beta \approx 1$ inasmuch as a and ϵ_e are likely to be only inaccurately known. It is possible to discriminate against the conversion electrons in the beta counter, by the use of appropriate absorbers, when the energy of the electrons is lower than that of the betas. High-energy conversion electrons arise from the conversion of high-energy gammas, but in such cases a is generally small.

If the beta and gamma have a directional correlation in the decay process, this must be known so that the observed coincidence rate for a given orientation of sample and counters may be properly interpreted. The use of high geometry in at least one detector tends to reduce effects of angular correlation.

Dead-time losses in the counters contribute errors in N that are generally small (of the order of 1 to 2%). If pulse-height selection is employed, as indicated in the schematic diagram (Figure 13), so that every pulse entering the coincidence circuit will also register in one of the singles channels, any losses that occurred at the detectors may be considered as part of the detection efficiencies. When the dead times appropriate to the singles channels are the same as those for coincidence registration (i.e., when the coincidence resolving time 2τ is negligible with respect to the dead times), the dead-time corrections cancel in the determination of N . However, this cancellation is no longer exact when the coincidence resolving time is comparable to the dead times of the singles channels.⁽⁷⁸⁾

Statistical analysis of errors must be applied to the data in order to estimate the accuracy. If careful measurements are made, the coincidence method for determining disintegration rates of nuclides with simple decay schemes may be expected to yield results having an error of 1-2%.

When complex decay schemes are involved, the treatment must be extended. For example, if a single beta is followed by more than one gamma ray, the gamma-counting efficiency is the sum of the counting efficiencies of the individual gammas with a small correction for γ - γ coincidences. If both gammas are converted, their conversion coefficients must be known and applied in the equations for N_β , N_γ , and N_c . When more than one beta ray is emitted, a series of coincidence measurements is made with varying absorber thicknesses in front of the beta counter. A coincidence absorption curve is obtained, which may be extrapolated to zero total absorber, and the disintegration rate is determined from a knowledge of the beta-branching ratios. Alternatively, energy discrimination in either

counter may be effected by the use of pulse-height analyzers. Other combinations of coincidence measurements (such as γ - γ , γ - e^- , or β - e^-) may also be employed in certain cases:

The importance of a number of the correction factors discussed above diminishes when the beta detector has high efficiency. Thus, e.g., with a 4π beta detector, the effects of source size, angular correlations, branching ratios, and conversion electrons are minimized. Systems utilizing $4\pi\beta$ detectors in a coincidence system have been developed with proportional counters⁽¹⁴³⁾ and with liquid scintillation counters.⁽¹⁴⁴⁾

Beta Counting of Liquid Samples

In general, counting of liquid samples is employed for relative measurements of rather energetic betas. As in counting of solid samples, the conditions of counting must be reproduced accurately for meaningful results. The problems of solution counting, such as the effects of type and energy of the radiation, geometrical arrangement, wall thickness of the counting tube, and the density and atomic number of the medium, are analogous to those encountered in counting solid samples. Experimental data on these effects in liquids, however, are not very extensive. Some of the problems are reviewed by Cook and Duncan,⁽⁷⁹⁾ Solomon and Estes,⁽⁸⁰⁾ Chiang and Willard,⁽⁸¹⁾ Walton,⁽⁸²⁾ and Friedman and Hume.⁽⁸³⁾

If absolute counting of liquids is necessary, it is recommended that the counter be standardized with a source of known disintegration rate.

Beta Scintillation Counting

Beta particles may be conveniently detected with scintillating crystals,⁽⁸⁴⁾ plastic scintillators (solid solutions of organic phosphors in plastics),⁽⁸⁵⁾ or liquid scintillator solutions.⁽⁸⁶⁾ These detectors must be suitably coupled to a photomultiplier tube for the detection and amplification of the light emitted by the scintillator, either by direct contact or by the use of light guides. A nonoverloading amplifier, pulse-height analyzer, and scaler complete the counting system. Such detectors offer the same advantages as proportional counters in the ability to discriminate between beta rays of different energy and in having small resolving-time losses. In addition, they may be used for beta rays of higher energy. In the low-energy region, however, proportional counters give somewhat better resolution.

The major problem in the use of scintillation detectors for low-energy radiation is the correction for background pulses from the thermal noise of the photomultiplier tubes. If the beta spectrum is sufficiently energetic, the background may be discriminated against by suitable biasing. Cooling of the photomultiplier tube, the use of two photomultipliers in a

coincidence arrangement,⁽⁸⁷⁾ or a combination of both techniques may be employed to reduce the background effect. In addition, techniques for increasing the light output of the scintillators may be utilized to improve the signal-to-noise ratio. For example, in liquid scintillator solutions dissolved oxygen quenches the light output,⁽⁸⁸⁾ and sweeping of the solution with an inert gas may be desirable. "Intermediate solvents,"⁽⁸⁹⁾ such as naphthalene or biphenyl, may be used as energy transferers in liquid scintillator solutions to combat the quenching action of some solutes, particularly of heavy elements. Since many materials exhibit a delayed fluorescence on exposure to ultraviolet light, caution should be exercised in the preparation of scintillation detectors. For example, m-xylene and benzene exhibit two periods of fluorescence⁽⁹⁰⁾ with half-lives in the region of 1-2 minutes and 15-20 minutes. The photocathode surface of the photomultiplier tubes should be protected from light or a sufficient period of time should be allowed after exposure for the decay of the fluorescence. Backgrounds, when critical, should, of course, be determined under conditions exactly reproducing those for sample counting.

A variety of counting techniques, employing scintillation detectors with external sources to take advantage of the opportunity for obtaining increased counting efficiency, has been investigated. Samples may be evaporated on the surface of a scintillating crystal or plastic and, to secure a 4π geometry, another crystal or plastic scintillator may be placed on top to form a sandwich^(95,164); the sample may be suspended on a thin foil⁽⁹¹⁾ or in the form of a fine powder⁽⁹²⁾ in a liquid scintillator medium or in a scintillating gel⁽⁹³⁾; and samples may be placed in a well drilled in a crystal or plastic scintillator.⁽⁹⁴⁾ None of these techniques, however, avoids the problem of sample self-absorption, and this must be treated in the same manner discussed under Geiger-Müller and proportional counting. For routine relative counting, in which reproducibility is the major criterion, these techniques may prove convenient and useful. This is particularly true for the well-counter, in which tubes containing solutions of the radioactive samples may be counted without further preparation (if the beta rays are of sufficient energy to penetrate the solution and container). For some applications, the suspension counting of low-energy or low-specific activity solid samples may be desirable. The 4π sandwich-crystal technique has been employed for absolute beta counting⁽⁹⁵⁾ and appears to be comparable with other techniques for medium and high-energy beta rays, but somewhat poorer for those of low energy. The higher efficiency of anthracene for X rays may make this technique advantageous for the assay of electron-capture nuclides.

The greatest advantage of scintillation counting for beta rays appears to lie in the opportunity of obtaining a 4π detector for low-energy and low-specific activity sources without problems of sample self-absorption. This may be achieved by incorporating the radioactivity into the molecules of the crystal itself⁽⁹⁶⁾ or by dissolving the radioactive sample in a liquid scintillator solvent.⁽⁹⁷⁾ In the first case, the applications are somewhat limited, and

the technique, in general, is not suitable or convenient for assay work. The prospects for liquid scintillation counting, however, seem very promising. Steyn⁽⁹⁸⁾ has shown that liquid scintillation counting may be successfully employed for the absolute standardization of beta sources. He has used refrigeration at -20°C to reduce the background. A 10-ml scintillator volume in a sample cell was placed directly on the face of the photomultiplier tube and covered with a hemispherical mirror. The voltage and amplification were adjusted so that more than half of the pulses saturated the non-overloading amplifier. Under these conditions, the differential distribution of the remaining pulses was essentially constant and the integral distribution curve was a straight line that could be extrapolated to zero pulse height to give the true disintegration rate of the source. Accuracies comparable with other methods seem readily attainable with this technique.

Most elements can be prepared in a form that is soluble in a liquid scintillator medium and which does not quench the light output of the scintillator seriously. Even aqueous samples may be made compatible with the organic solutions by the use of alcohol or dioxane. Solutions of the sources may be conveniently measured and introduced into the scintillator, and a wide range of energies and sample weights may be accommodated by the adjustment of scintillator volumes. This technique, although relatively new and in the process of further development, appears to offer a satisfactory solution to the problem of the absolute assay of low-specific activity and/or low-energy beta sources.⁽¹⁴⁵⁻¹⁴⁸⁾

ALPHA COUNTING*

The extremely short range of alpha particles in matter necessitates the use of detectors with thin windows or provisions for the introduction of samples into the active volume (internal counters). For general alpha-assay work, pulse ionization chambers or proportional counters have proven most satisfactory. Current-measuring devices have a number of disadvantages, such as the dependence of ionization current on the alpha energy and the sample thickness, the lack of distinction between the ionization due to alphas and that due to β , γ , or cosmic radiation, and the inability to measure very weak sources. In pulse chambers one alpha will give rise to a single pulse over a wide range of energies. Sample self-absorption is not so important, since a pulse may be detected even after a large fraction of the range has been expended. An alpha pulse can be discriminated from β , γ , or cosmic ray pulses, and extremely low backgrounds (<1 count per minute) can be attained, thus permitting the accurate measurement of very weak samples.

In general, the same considerations discussed in the case of the determination of beta disintegration rates apply to alpha determinations with quantitative rather than qualitative differences. The counting yield is again dependent on such factors as geometry, scattering, absorption, and detection probability.

Geometry Factor. Two geometry factors are generally used: "50%" (or 2π) and "low" ($<1\%$). In the 50% chamber the source is placed inside the chamber and, since half the particles are emitted away from the plate, the relative solid angle is 0.5. In the low-geometry counter the source is placed at a known distance from an aperture that opens into the chamber. The relative solid angle is calculated from the known dimensions of the system (see Appendix I).

Scattering. Since alpha particles do not scatter easily, the scattering factor may be considered as unity, even in low-geometry chambers. However, backscattering from the sample plate does occur and must be corrected for in 50% chambers.

Backscattering. Not much experimental data have been accumulated on the variation of alpha backscattering with the alpha energy and atomic number of the scatterer. However, it is known qualitatively that backscattering increases with increasing atomic number Z and with decreasing alpha energy. For the most commonly used sample plates (Pt), the backscattering factor has been determined as 1.04 in a 50% chamber.^(99,100) This result is in good agreement with theoretical predictions.⁽¹⁰¹⁾ In low-geometry chambers the backscattering effect is negligible, and this instrument is useful as an absolute geometry standard.

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* This section is based largely on the excellent and comprehensive discussion of alpha assay by A. H. Jaffey.⁽³⁾

Absorption. In alpha assay work it is the general practice to count under such conditions that the effect of external absorbers is negligible. Thus internal counters or low-geometry chambers with a total absorber path (in gas plus window) equal to less than 90% of the alpha range are used.

For some measurements not requiring high precision, it is possible to use a foil-absorption technique to separate the contributions of mixtures of alphas having different ranges. The sample is mounted in a 50% geometry chamber, and the counting rate is measured as a function of the absorber thickness placed over the sample. The alphas are absorbed linearly, and the "peeling-off" process is used to determine the contributions of the individual components (Figure 14).

Figure 14

Resolution of a complex curve (range measurement by foil absorption in a 50% geometry pulse chamber) into two alpha-emitting components with extrapolated ranges of 3.18 and 4.45 mg/cm^2 , respectively.

Thin smooth samples are necessary for the application of the method, since excessive self-absorption in the sample results in a nonlinear absorption curve that might erroneously be interpreted as indicating more than one alpha (Figure 15). Other methods, e.g., the use of a differential range chamber⁽³⁾ or pulse analysis, may also be utilized for the purpose of resolving a multicomponent alpha source, but they require special instrumentation.

Figure 15

Effect of excessive self-absorption due to sample thickness. Curve A shows the type of curve determined from a thin smooth sample. Curve B shows the type of curve determined from a sample having the same type of radioactivity and the same counting rate with no absorber but with an excessive amount of foreign material, e.g., carrier salt.

Self-absorption.^(3,127-132) If visible amounts of solid are present in an alpha sample, losses in counting rate in a 50% geometry chamber may

be expected because of self-absorption. In a smoothly spread sample the fraction F of alphas lost by self-absorption has been calculated⁽¹⁰²⁾ as*

$$F = \frac{1}{2}t/R \quad ,$$

where t is the sample thickness and R is the effective range of the alpha particle (i.e., the maximum distance an alpha particle can travel through the source material and still register a pulse in the chamber). The effective range R is always less than the true range. Since it is difficult to prepare uniformly spread samples, the effect of sample thickness is generally larger than calculated.

Scratches in the surface of the mounting plate may also cause losses in counting rate because of absorption of alphas emitted at low angles from the part of the sample deposited in the scratch.

In low-geometry chambers, only those alphas emitted nearly vertically are effective, and, if the sample thickness (plus gas and window thickness) is less than the range of the alpha, no appreciable losses will occur. For thicknesses greater than R , the fraction lost will be $F = 1 - (R/t)$. Clumping of the sample will usually make the effective thickness larger than the average thickness of the source.

Self-absorption correction curves may be determined by methods analogous to those employed in the case of beta particles. In the determination of such self-absorption curves, it should be realized that as the thickness of sample increases the plateau changes, and it is advisable to extrapolate the plateau for each sample to zero pulse height for comparison.

Plateaus. Although a knowledge of the proper operating conditions for the detecting instrument is assumed for the purposes of this report, a discussion of the proper operating voltage in relation to the sample properties is pertinent. Thus it may not always be desirable to use a counter at the flattest portion of its plateau. For the comparison of samples having various degrees of energy degradation (e.g., due to different self-absorption in the sources), it may be desirable to count far up on the plateau, where the degraded pulses will register. For thin samples on the same backing, reproducible counting can be obtained at any point along the plateau, although it is generally desirable to operate a short distance above the knee of the plateau. In internal proportional counters the counting efficiency may depend on the sample area for large samples when operating just above the plateau knee, and it is advisable to keep the samples confined to areas 1 cm in diameter or less.

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* This formula applies to thicknesses less than R . For samples of greater thickness $F = 1 - (R/2t)$.

Beta pulse pile-up. The proportional alpha counter may be used to count alpha particles in the presence of betas, since the pulse heights produced by the betas are much smaller and may be discriminated against by appropriate biasing. However, when the number of beta particles becomes great enough so that they pile up to produce pulses of sufficient magnitude to be registered, the alpha plateau is shortened and the input threshold of the instrument is affected in such a way that a net reduction in alpha counting rate may be observed.⁽¹⁰³⁾ The magnitude of the reduction depends on the operating voltage, beta intensity, beta energy, alpha pulse-amplitude distribution, input amplitude threshold, and individual instrument-circuit characteristics. For a beta intensity of 3×10^7 disintegrations per minute (dpm), the reduction is about 0.5%, and for 4×10^8 dpm, it is about 2%. With sufficient beta activity, the plateau disappears and alpha counting is impossible.

Counting standards. The reproducibility of counting efficiency is normally checked by the use of alpha-counting standards. If it is desired to adjust the external or internal gain so that a previous counting rate of the standard is reproduced, a thick sample should be used as a standard. In such a sample a wide and continuous distribution of pulse heights is produced, and, since essentially no plateau exists, the counting rate is very sensitive to small variations in gain and may be accurately and easily reproduced. For counting a series of similar samples over a period of time, it is usual to measure ratios of the counting rates of sample and a standard. These ratios are generally more reproducible than the individual counting rates.

Resolution losses. The corrections for coincidence or resolution losses are discussed in Appendix II. The low-geometry counter is suggested for very active samples when the resolution loss correction in a 50% chamber is too large to be made accurately.

Absolute counting.^(3,133,149) Absolute disintegration rates may be determined in 50% chambers when the backscattering correction is known and self-absorption in the source is negligible. The counting efficiency, however, will vary along the plateau because of the slope resulting from the backscattering effect and variations of internal amplification for tracks in different parts of the chamber. These effects limit the accuracy of such absolute determinations and introduce errors of the order of 1%.

Low-geometry counters avoid these effects, and very accurate disintegration rate determinations can be made, provided the geometry is accurately defined and calculated or the counter efficiency is calibrated with a source of known disintegration rate.

The application of liquid scintillation counting to alpha assay avoids the problem of self-absorption in the source material since the sample is homogeneously dispersed in the counting medium. Both differential and

integral measurements may be made, and the technique has been shown to be essentially identical in accuracy with low and intermediate-geometry counters.(150-152)

Relative counting. For many purposes the relative counting rates of sources is sufficient. It is then only necessary to reproduce the counting conditions (such as backscattering and self-absorption) by mounting the samples identically. It is difficult to reproduce self-absorption accurately except by making it very small. For sample thicknesses of the order of 0.1 mg/cm^2 , it is usually possible to spread the sample sufficiently uniformly to attain reproducibilities to within a few per cent.

GAMMA COUNTING

Relative Counting

The high penetrability of gamma radiation in matter permits some simplification of relative-counting techniques. The problems of absorption and sample thickness, which are extremely important in alpha and beta counting, are minimized in gamma counting. Thus, relative measurements of gamma intensity are particularly useful for samples that differ in chemical composition or size and for liquid samples. Of course, in measuring gamma radiation, sufficient absorber must be placed in front of the detector to screen any accompanying beta particles, and such an absorber should be of low-Z material in order to minimize the bremsstrahlung production that accompanies the stopping of the betas.

It is sometimes difficult to maintain a constant geometric relationship between source and detector when solid samples of different size are being compared. Such comparisons, however, may readily be made in solution, using, e.g., a high-pressure ionization chamber detector.⁽¹⁰⁴⁾

Absolute Gamma Counting

In the determination of absolute intensities of gamma radiation the major problem is a knowledge of the intrinsic efficiency of the detector. This efficiency is dependent on the energy of the gamma ray and the nature of the material of which the detector is made. The ordinary brass-walled, argon-filled, end-window Geiger counters have an efficiency of about 1% per Mev of gamma energy. This relationship is quite linear above ~ 0.1 Mev. Below this energy the efficiency decreases less rapidly with decreasing energy and finally rises again because of the significant absorption in the gas filling. Many devices have been used to increase the efficiency of detectors for gamma radiation, e.g., multiple chamber counters, counters lined with materials of high atomic number, high-pressure ion-chambers, and scintillation detectors.

Any of these detectors may be calibrated with sources of known disintegration rate, and an efficiency curve as a function of gamma ray energy may be determined. The decay schemes of the standards used must, of course, be known, so that the number of gamma rays accompanying each disintegration may be calculated. The disintegration rate may be determined by using the methods outlined in the section on beta counting. The intensity of gamma radiation of known energy in a source is then determined from the counting rate of the sample and the efficiency as read from the calibration curve.

In many instances, two or more gamma rays are involved in a decay process. If the energies are sufficiently different, the relative contributions

may be determined from an absorption curve (usually in lead). The logarithm of the activity is plotted against the thickness of absorber. Since gamma radiation is exponentially absorbed, each component will exhibit a half-thickness characteristic of its energy. The resolution of this method is rather poor, however, and, in general, the energies must differ by about a factor of two for a successful analysis of the curve. (The resolution is somewhat better in the region of a few hundred kev where the absorption coefficient is a more sensitive function of the energy.) For energies that do not differ sufficiently, a single exponential curve with a half-thickness characteristic of the average energy of the components is obtained.

Experimental verification of theoretical absorption coefficients may be obtained under conditions of so-called "good" geometry, i.e., collimated beams with large distances separating source, absorber, and detector so that secondary radiation is not recorded.⁽¹⁰⁵⁾ Under the usual laboratory counting conditions, "good" geometry is rarely attained, and secondaries and scattered radiation may be detected. The effect on absorption curves is to alter the strictly exponential character of the absorption and to introduce curvature that may be erroneously interpreted as an additional component. The exact nature of the effect is dependent on the particular conditions involved. In a satisfactory technique, one lead absorber ($\sim 1 \text{ gm/cm}^2$) is placed adjacent to the detector window. The position of additional absorbers between the sample and the first absorber is not critical. Such an arrangement results in exponential absorption, although the half-thicknesses observed are somewhat larger than theoretical. The efficiency curve should be determined with the same absorber in position adjacent to the detector if absolute intensities are to be determined.

Scintillation Counting - Pulse-height Analysis

Scintillation counting has proved extremely valuable for gamma counting,⁽¹⁰⁶⁾ and it is rapidly replacing other techniques. By the use of thallium-activated NaI crystals, detection efficiencies approaching 100% may be achieved. The scintillation detector may be calibrated by using standard sources and may be used for integral or differential counting. Integral counting is perhaps somewhat simpler and is not so subject to the electronic difficulties associated with pulse-height analysis. It is recommended for relative measurements or for absolute intensity measurements of samples known to emit a single gamma or X ray. (Multiple gammas may be separated by absorption, as previously described, with the same limitation on resolution.) For absolute measurements, one must know the solid angle between the source and the entering face of the crystal, and also an efficiency factor which expresses the probability that a gamma quantum, after striking the crystal, produces a light pulse large enough to register over the background of noise.

This "total intrinsic efficiency," as it has been called, depends upon the energy of the gamma quantum, the size and shape of the crystal, and the distance of the source from the crystal, inasmuch as the source distance affects the paths of the gamma rays in the sodium iodide. Computations have been made of the total intrinsic efficiency by using appropriate theoretical gamma-ray absorption coefficients for NaI, and results have been given by Bell, Davis, and Lazar* and more extensively by Wolicki, Jastrow, and Brooks.⁽¹⁰⁷⁾ Figures 16 to 24 give the results of Bell, Davis, and Lazar for $1\frac{1}{2} \times 1$ -inch crystals, 3×3 -inch crystals, and 3×3 -inch crystals with the edge of the entering face beveled off to reduce the number of small pulses arising from short path lengths near the corner.

A single, or (better still) a multichannel pulse-height analyzer for use with the sodium iodide crystals ensures a much more powerful method for gamma-ray measurements.⁽¹⁰⁷⁾ Not only can sources that emit gamma rays of more than one energy be dealt with, but more reliable results in absolute measurements can often be obtained.^(112,113,116,117) One reason for the increased reliability lies in the fact that the pulse-height spectra consist in general of two parts: the pulse distribution from gamma rays that lose all of their energy in the crystal, and the pulse distribution from gamma rays that lose only part of their energy in the crystal. The first part stands as a separate peak that is especially prominent in the larger crystals, and use of the area under this peak as a measure of the gamma-ray intensity insures against the measurement of scattered (and therefore degraded) gamma rays. The requirements of source surroundings then become less stringent. To do this conveniently, a figure for the "intrinsic peak efficiency" is needed, that is, the probability that a gamma quantum, after striking the entrance face, loses all of its energy in the sodium iodide. Figures for intrinsic peak efficiencies have been obtained experimentally in relation to the calculated total intrinsic efficiencies by comparing, in a scatter-free geometry, the intensities of the total-energy peaks with the total intensities of the scintillation spectra. Work of this kind has been carried out by Kalkstein and Hollander,⁽¹¹¹⁾ by Heath and Schroeder,⁽¹¹²⁾ and by Lazar, Davis, and Bell.^(108,109,110) The curves obtained by Lazar et al. are reproduced in Figures 25 and 26. The curves have been checked by comparison with independently calibrated gamma-ray sources and are thought to be accurate to within five per cent. To use them in an absolute gamma count, the area under the total-energy peak, as given by the scintillation spectrometer, is measured and divided by the intrinsic peak efficiency read from Figure 25 or 26 for the appropriate crystal size, source distance, and gamma-ray energy. Finally, the solid angle factor between source and crystal is applied. In all of this work it is assumed that the source is small compared to the dimensions of the crystal and that it is situated on a prolongation of the axis of a cylindrical (perhaps beveled) crystal.

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*Unpublished.

Figure 19

Total intrinsic efficiency of NaI(Tl) crystal 3-in dia x 3-in high.
 Source located on crystal axis h-cm above crystal
 h = 3 to 70 cm

The properties of some useful standards for calibration of systems not adequately represented by the curves of Figures 16 through 26 are given in Table I.^(113,114) Disintegration rates for these nuclides may be determined by 4π or coincidence-counting techniques.

Table I
USEFUL STANDARD SOURCES FOR γ -INTENSITY CALIBRATIONS

Nuclide	Half-life	γ Energy (Mev)	Number of Gammas per Disintegration	β Energy (Mev)	Method of Determination of Disintegration Rate
Na ²²	2.60 yr	1.277 0.511	1.11 ^a 2.00 ^a	0.542	4π counting, β - γ coinc.
Na ²⁴	15.0 hr	1.38 2.76	1.00 1.00	1.39	4π counting, β - γ coinc.
Sc ⁴⁶	85 d	0.89 1.12	0.995 1.00	0.36	4π counting, β - γ coinc.
Co ⁶⁰	5.27 yr	1.332 1.172	1.00 1.00	0.306	4π counting, β - γ coinc.
Nb ⁹⁵	35 d	0.745	1.00	0.160	4π , β - γ coinc.
I ¹³¹	8.14 d	0.364	0.80	0.815	4π
Au ¹⁹⁸	2.69 d	0.412	0.957	0.963	4π
Hg ²⁰³	47 d	0.279	0.81	0.208	4π , β - γ coinc.
Tl ²⁰⁸	3.1 min ^b	2.62	1.00	1.79	α -counting
Am ²⁴¹	500 yr	0.0596	0.357		α -counting

^aNumber of gammas per positron.

^bSupported by 10.6-hr Pb²¹²

Convenient standards may not be readily available for calibration, particularly in the low-energy region. In the energy region below about 150 kev, essentially all of the gamma rays interact by the photoelectric process, and the pulse-height distribution exhibits a full-energy or photopeak. Events occurring near the edges of the crystal may result in the loss of pulses from the full-energy peak. To avoid these edge effects, the crystal edges may be screened by the use of a collimator or iris. Under these conditions, the intrinsic efficiency of the crystal is essentially 100% and the geometric efficiency may be calculated from the dimensions of the system.

Iodine X rays produced near the surface of the crystal may not interact further and may leave the crystal. This loss is observable in the pulse-height distribution as an additional peak about 28 kev below the photopeak for incident gamma-ray energies of less than about 150 kev (and

greater than 33.2 keV, the K-edge of iodine). At higher incident energies the resolution of scintillation spectrometers is insufficient to distinguish a separate escape-peak. The contribution of pulses in the escape-peak must be added to those in the photopeak to give the total gamma intensity. The escape-peak correction may be calculated or determined empirically. Axel⁽¹¹⁵⁾ has reviewed the problem and presents calculations of the escape-peak probability as a function of gamma-ray energy based on the approximations of infinitely wide and thick crystals and interaction involving only the photoelectric process. Both assumptions are shown to be valid for the energy range (<150 keV) considered. Figure 27 illustrates the results for the cases of "good" geometry (gammas incident normal to plane of crystal), "intermediate" geometry (crystal subtends a cone of half angle equal to 60°), and "poor" geometry (half-angle of cone equal to 90°, i.e., source in direct contact with crystal).

Figure 27

X-ray escape probability for infinitely thick, infinitely wide NaI crystal. (Gamma rays should produce X rays more than 2.5 mm from the edges or the back surface of the crystal.)

The defined geometry technique is not convenient to apply at higher incident gamma-ray energies since the collimating screens become too thick and may introduce scattering effects from their edges. In addition, at higher gamma-ray energies the Compton effect becomes appreciable, and the scattered gammas may undergo multiple processes that contribute to pulses in the photopeak. Such effects are extremely difficult to calculate, and calibration techniques using standard sources are advisable for these energies.

In those cases in which two or more gamma rays are emitted within the resolving time of the instrument, a peak representing the sum of the gamma-ray energies may be observed. The probability of observing such a peak increases with increasing counting efficiency of the system. For

absolute counting rate determination such events must be added to the counting rate of the gamma ray of interest to give the total counting rate.

The technique of gamma-ray spectrometry utilizing multichannel pulse-height analysis is an extremely useful tool for the assay of radioactive nuclides.⁽¹⁵³⁻¹⁵⁵⁾ The efficiency of the method is determined by means of calibrated sources, and direct assay of mixtures of radionuclides may be carried out in many cases without chemical separation.

The Compton distribution of gamma rays of higher energy must be subtracted from the photopeaks of the less energetic gamma rays, and a number of systems for accomplishing this have been suggested. A "complement-subtraction method"⁽¹⁵⁶⁾ makes use of a feature of some multichannel analyzers which permits the display of an inverse spectrum.⁽¹⁵⁷⁾ When a calibrated source of a particular nuclide is then counted in the normal mode of operation, its spectrum will be subtracted out. The number of counts required to completely erase this portion of the complemented (or inverse) spectrum gives the abundance of the nuclide of interest in the original mixture. The Compton distribution may be minimized by the use of large crystals⁽¹⁵⁸⁾ or, even better, by anticoincidence rejection of the radiation escaping the crystal.⁽¹⁵⁹⁻¹⁶²⁾

Catalogs of gamma-ray spectra have been compiled^(153,163) and are useful aids in the identification of unknown radioactive samples.

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APPENDIX I. FORMULAS FOR GEOMETRY CALCULATIONS*

The "geometry" of the aperture for a point source, G_p , is defined as the fraction of the total solid angle subtended by the aperture, i.e.,

$$G_p = \Omega_p / 4\pi \quad (1)$$

For the case of a point source on the axis of a circular aperture (see Figure A1),

$$G_{p'} = \frac{1}{2} \left[1 - \frac{Z}{D} \right] = \frac{1}{2} \left[\frac{a^2}{D(D+Z)} \right] \quad (2)$$

Figure A1

Relation of source and aperture. O = origin,
 $PQ = R$, $OP' = Z$, $PP' = \rho$, $\cos \gamma = Z/R$,
 $OP = \ell = (Z^2 + \rho^2)^{1/2}$, $Z/\ell = \cos \theta = x$,
 $D = (Z^2 + a^2)^{1/2}$

*For a much more extensive discussion and tables of point source geometries, see A. H. Jaffey, Rev. Sci. Instr. 25, 349 (1954). For tables of solid angles subtended by circular discs and cylinders, see A. V. H. Masket, R. L. Macklin, and H. W. Schmitt, ORNL-2170 (Nov. 1956), and for a discussion thereof see A. V. Masket, Rev. Sci. Instruments 28, 191 (1957).

For the case of a point source off the axis of a circular aperture, and $\rho/D < 1$,

$$\begin{aligned} G_p &= G_{p'} - \frac{3}{8} \rho^2 \frac{a^2 Z}{D^5} + \frac{15}{32} \rho^4 \frac{a^2 Z}{D^9} \left(Z^2 - \frac{3}{4} a^2 \right) + \dots \\ &= G_{p'} + \frac{1}{2} \frac{Z}{D} \sum_{n=1}^{\infty} \frac{(-1)^n (2n+1)!}{2^{4n} (n!)^2} \left(\frac{\rho^2}{D^2} \right)^n \sum_{k=0}^n m_k, \end{aligned} \quad (3)$$

where

$$m_k = \frac{(-1)^{k+1} (4n-2k)!}{(2n-k)! (2n-2k+1)!} \left(\frac{Z^2}{D^2} \right)^{n-k}$$

and $G_{p'}$ is taken for the same value of Z and is given by equation (2).

For the case of a uniformly spread circular source of radius b , coaxial with and parallel to the aperture,

$$G_s = G_{p'} - \frac{3}{16} b^2 \frac{a^2 Z}{D^5} + \frac{5}{32} b^4 \frac{a^2 Z}{D^9} \left(Z^2 - \frac{3}{4} a^2 \right) + \dots \quad (4)$$

Approximate Formulas

For small displacements the geometry of an off-axis point may often be fairly well approximated by $G_{p'}$. Likewise, when b is sufficiently small, the geometry of a uniformly spread source may be approximated by $G_{p'}$. The error introduced in these approximations may be determined from the following:

For the case of small ρ/D or b/D , approximately,

$$G_p = G_{p'} \left\{ 1 - \frac{3}{4} \rho^2 \left[\frac{Z(Z+D)}{D^4} \right] \right\} \quad (5)$$

and

$$G_s = G_{p'} \left\{ 1 - \frac{3}{8} b^2 \left[\frac{Z(Z+D)}{D^4} \right] \right\} \quad (6)$$

If a/Z is also small,

$$G_p = G_{p'} \left(1 - \frac{3}{2} \frac{\rho^2}{Z^2} \right) \quad (7)$$

APPENDIX II. CORRECTIONS FOR COUNTER NONLINEARITY

The instruments used to detect radioactivity have finite resolving powers, and two events that occur within a short time interval may not be distinguished and recorded separately. Thus some counts may be missed, the fraction lost increasing with increasing counting rate of the sample.

The nonlinearity (sometimes referred to as "resolution loss," "dead-time loss," or "coincidence loss") may be determined in a number of ways:

(1) the method of paired sources, (2) measurement of calibrated samples, (3) measurement of multiple aliquots, and (4) measurement of the decay of a sample of known half-life. Electronic methods of determining the dead-time may also be used.

1. The Method of Paired Sources

Each of two radioactive sources of approximately equal strength is measured separately and then together. Backgrounds must be subtracted from each measurement, and care must be taken to ensure constant geometry factors for the measurements. If nonlinearity is present, the counting rate of the combined samples will be less than the sum of the individual counting rates. It is advisable to prepare the sources on sturdy material and to support them in a reproducible fashion, e.g., as illustrated in Figure A2. The sources should be small enough so that they can lie side by side, without touching each other, on the support. If the sources are too far apart, effects of the variation of counter sensitivity along the window diameter may be encountered.

Figure A2

Suggested source-mount for paired-sample technique. M is a metal plate holder that fits shelf of counter assembly, A and B are sources mounted on metal slides, and C is a metal clip to hold slides in position. Blank slides are used for backgrounds and to replace either sample during separate counting rate determinations.

The sequence of measurements necessary for the evaluation of the resolving time is as follows:

- a. Place both blanks in position and measure the background b .
- b. Replace one blank by sample A and measure counting rate R_A .
- c. Replace other blank by sample B (taking care not to disturb position of sample A) and measure the combined counting rates R_C .
- d. Replace sample A by blank (taking care not to disturb the position of sample B) and measure counting rate R_B . τ is then given by

$$\tau = \frac{R_A + R_B - R_C - b}{R_C^2 - R_A^2 - R_B^2}$$

The highest statistical accuracy for a given total expenditure of time is obtained when

$$t_A : t_B : t_C : t_b = \sqrt{R_A} : \sqrt{R_B} : \sqrt{R_C} : \sqrt{b} \approx 1 : 1 : \sqrt{2} : \sqrt{b/R_A}$$

It is convenient and desirable to count R_A , R_B , and R_C for equal periods of time to obtain the highest statistical accuracy for a given total number of counts. It is also desirable to make t_b somewhat greater than indicated by the foregoing ratio.

If a simple equation involving only one parameter relates the true and recorded counting rates, only a single set of measurements involving one pair of samples is necessary. An example of such an equation is

$$N = R + \tau R^2$$

where N is the true counting rate, R is the observed counting rate, and τ is the resolution-correction parameter or dead-time.

For correction formulas involving two or more parameters, more data are necessary and may be obtained by measuring a series of paired sources of different counting rates.

A complete treatment of the method of paired sources is given by T. P. Kohman in Seaborg, Katz, and Manning, The Transuranium Elements, N.N.E.S. Vol. 14B, Div. IV, Paper 22.50 (McGraw-Hill, 1949), and Anal. Chem. 21, 358 (1949).

2. Measurement of Calibrated Samples

If the resolution loss correction has been determined on one counter, a sample or series of samples of different counting rates can be counted with this counter and the true counting rates determined. The samples may then be used to determine the resolution loss corrections for other counters.

3. Measurement of Multiple Aliquots

A series of samples may be prepared from a stock solution, each differing in activity by a known multiple from the others. These can then be counted, and the resolution losses may be determined from the decrease in the expected counting rate at higher activities. Of course, careful aliquoting and plating of samples are imperative for the application of this technique.

4. Measurement of the Decay of a Sample of Known Half-life

This technique, in principle, is equivalent to method 3 above. The decay of a pure radionuclide with accurately known half-life is followed until the activity reaches a level at which the resolution losses are negligible and the known half-life is observed. The deviation of the initial portion of the decay curve (high counting rate) from the expected half-life gives a measure of the resolution losses.

In the application of all of these methods, small differences between large numbers are generally encountered. Thus it is necessary that counting rates be determined with high statistical accuracy. Methods 2,3, and 4 are somewhat more convenient, since they give correction curves that can readily be applied to counting data.