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Foreword

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Subcommittee on Radiochemistry

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I. ABSOLUTE MEASUREMENT OF ALPHA EMISSION

A. INTRODUCTION

The discovery of radioactivity by Becquerel in 1896 and the isolation of ^{226}Ra and ^{210}Po from pitchblende by Marie Curie in 1898 stimulated research by many investigators into the nature of the rays emitted by radioactive substances. By 1910 alpha rays had been shown to be helium atoms with a double positive charge, the mass to charge ratio had been measured, and the velocities and ranges in air of the alpha rays from radium and most of its decay products had been measured. In addition, many of the elements in the uranium decay chain had been identified and their half-lives measured.

In 1908, Regener[1] devised a scintillation method of counting individual alpha particles and made the first measurements of the alpha emission rate of a source by alpha scintillation counting. He arranged a glass plate thinly coated with finely divided zinc sulfide particles so that the alpha particles impinged on the fluorescent surface of the plate. He then counted the individual scintillations visually using a microscope with a magnification of about 30. The scintillation of ZnS and some other substances by alpha ray bombardment had been observed and studied by other scientists several years earlier. In a subsequent paper (in 1909)[2], Regener described the use of his scintillation method to measure the alpha disintegration rate of a ^{210}Po preparation.

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At about the same time Rutherford and Geiger[3] made the first proportional counter and measured the alpha emission rate of a RaC (^{214}Bi) source by detecting the alpha particles individually. In the experimental arrangement, a thin, small area source of ^{214}Bi was placed in an evacuated tube several meters from a small mica-covered aperture on the end of a cylindrical counting chamber. The counting geometry was known by calculation from the aperture area and distance from the radioactive source. It was made intentionally low (about 10^{-8}) to have the counting rate in the range of three to five per minute. The counting chamber was about 1.8 cm in diameter by 25 cm long, with a central high voltage wire. By operating the chamber at a pressure of about 40 mm of mercury and an applied voltage of about 1200 (just below the sparking voltage), each alpha particle entering the chamber caused an easily visible throw of a quadrant electrometer needle. Beta particle events gave only a slight pulse and were easily rejected. With this counting arrangement, Rutherford and Geiger determined the alpha disintegration rate of the sample of RaC which in turn was calibrated in terms of curies by comparing its gamma activity with that of a standard ^{226}Ra source.

Rutherford and Geiger also counted their sources by the scintillation method of Regener and obtained good agreement between the two methods. They used the same low geometry arrangement as in the electrical method but with a ZnS screen replacing the counting chamber. Their apparatus was the forerunner of the low geometry alpha scintillation counters now used for calibrating alpha sources. The only basic difference in the modern apparatus is the substitution of a multiplier phototube and electronic counting circuits for the human eye and microscope.

During the decade following the development of the electrical and scintillation methods for counting alpha particles, various investigators made repeated measurements of the alpha emission rates of many of the alpha active radionuclides found in nature. In 1924, Geiger and Werner[4] devised a coincidence method for determining the absolute number of alpha particles impinging per unit of time on the scintillation screen of their counter. They focused two microscopes on the screen so that two observers could simultaneously count the scintillations. The observers independently recorded the counts seen, as they occurred, making marks on a moving tape so that coincident counts as well as the total counts of each observer could be later read. The mathematical analysis by Geiger and Werner of the coincidence counting data is identical to that now widely used for determining

absolute disintegration rates by coincidence counting. If N is the true number of scintillations that occurred on the screen, and λ_1 and λ_2 are the counting efficiencies of observer 1 and observer 2, respectively, then

$$N_1 = \lambda_1 N$$

$$N_2 = \lambda_2 N$$

$$\text{and } C = \lambda_1 \lambda_2 N$$

Where N_1 and N_2 are the observed counts by observer 1 and observer 2, respectively, and C is the number of coincident counts. Therefore, one can calculate N from the recorded data, N_1, N_2, and C, using

$$N = \frac{N_1 N_2}{C}$$

The assumptions made are that each observer's counting efficiency is constant, that the number of chance coincidences are negligible, and that there are no scintillations which cannot be seen by at least one of the observers.

The emission of penetrating rays by uranium was first detected by the darkening of photographic plates and many of the subsequent studies of radioactive materials were made by using photographic plates to detect the alpha, beta and gamma rays. In 1910, Kinoshita[5] published the results of his studies of the effect of alpha rays on photographic emulsions of two types, an ordinary photographic emulsion and a high density emulsion. The latter was shown to be best suited for radiography. He attempted to relate the optical density of the developed film to the amount of alpha exposure but found the reproducibility to be rather poor. However, he found that the individual grains of silver developed along an alpha path through the emulsion could be counted under a microscope.

Since he had arranged the sources so the alpha rays impinged normal to the emulsion and since the emulsions were thin, only one or two grains were developed per alpha. Therefore, the grain count gave only an approximate count of the number of alpha events. The limitation in using the photographic method for quantitative alpha counting at that time was in the lack of thick, high-density emulsions, since only alphas entering at an oblique angle would leave an unambiguous track. The first thick emulsions (about 50 microns) were not prepared until about 1927.

B. CHARACTERISTICS OF ALPHA RADIATION

Hyde, Perlman, and Seaborg[6] list the alpha spectra of over 200 known alpha emitting isotopes, including those that decay by other modes besides alpha emission. Their half-lives range from a fraction of a second to 1.4×10^{10} years. A recent and convenient summary of radioisotope data is available in the book, "Table of Isotopes"[7]. Alpha particle energies range from 2.5 to nearly 12 MeV, including the long-range alphas that originate from excited nuclear states. Most of the alpha emitting isotopes in common use have alpha particle energies ranging from 4 to 6 MeV.

The range in air of the ^{232}Th alphas (3.95 MeV) is 2.4 cm, and that of the alphas from ^{239}Pu, ^{240}Pu, ^{232}U, ^{210}Po, and others having an energy of about 5.2 MeV is 3.7 cm. A 6 MeV alpha has a range of about 4.6 cm. A useful empirical formula[8] for estimating the range of 4 to 6 MeV alpha particles in air at standard conditions is:

$$R = 0.309\, E^{3/2},$$

in which R is in centimeters and E is in MeV. Ranges of particles in other substances are generally expressed in units of milligrams per square centimeter. One of several empirical formulas[8] for estimating the range of alphas in materials other than air is:

$$R_A\ (mg/cm^2) = 0.56RA^{1/3}$$

in which A is the atomic weight of the material and R is the range in air in centimeters. To calculate the range in centimeters, divide R_A by $10^3\,\rho$, where ρ is the density of the material in g/cm^3. Many sets of experimental range-energy curves have been published, the most complete being those for alpha particles and protons in Al, Cu, Ag, Pb, photographic emulsions, and several organic materials. A good theoretical treatment of the range-energy relation has been given by Bethe and Ashkin[9].

Moving charged particles with energies large compared to those of the bound electrons of an absorber lose energy to the absorber mainly by excitation and ionization of the absorber atoms. Livingston and Bethe[10] give a theoretical quantum mechanical relationship that predicts the energy loss of charged particles due to collision processes in an absorber:

$$-\frac{dE}{dx} \propto NZ\left(\frac{ze}{v}\right)^2$$

Where $-\frac{dE}{dx}$ is the energy loss per centimeter, ze and v are the charge

and velocity of the moving particle, N is the number of absorber atoms per cm^3, and Z is the atomic number of the absorber. The equation predicts correctly that the number of ions formed per centimeter by the particle increases almost exponentially as the particle slows down. The equation breaks down at the tail of the alpha track, when the alpha has slowed down to much less than 1 MeV.

The initial velocity of a 5.3 MeV alpha particle is about 1.5×10^9 cm/sec. For such a particle, the average energy loss per ion pair formed in air is 35.6 eV, and the number of ions formed along the total alpha track in air is about 1.5×10^5. The size of this number influences the size of the voltage pulse from an ionization chamber. The range of the particle and the density of ions along the trajectory of the particle can influence the design of an ionization chamber. For example, fission product ions have very short ranges in an absorber and have very large initial charges. However, the charge decreases along the ion path by capture of electrons. As a consequence, the specific ionization of a fission particle is highest near the beginning of its trajectory and a proportional counting chamber can be made efficient for fission particles while discriminating against alpha particles by keeping the chamber dimensions small enough so that the alpha particles do not expend all of their energy in the sensitive volume.

C. METHODS OF ABSOLUTE ALPHA COUNTING

1. Introduction

In most analyses for alpha emitting isotopes in chemical and biological work, relative methods for measuring alpha activity are sufficient. By this we mean that the amounts of alpha activity collected or recovered in the process are compared with the amounts taken initially. The counting efficiency is simply kept constant by reproducing the sample mounting and counting techniques to a degree controlled by the degree of accuracy desired. However, on some occasions an absolute measurement of alpha activity is needed. In such a case, either an absolute or a calibrated counting method is required, and in the latter case, an absolute method of counting may be needed to standardize the method and the alpha counter. Typical applications in which absolute or calibrated alpha counting methods may be required are measuring the amount of an alpha active radionuclide in ores, minerals, soils, water, etc.; determining the amount of an alpha emitter in biological specimens; measuring the inputs and outputs of a process; and measuring yields from a fission or activation process.

Earlier general reviews of absolute alpha counting are found in books by Seaborg and Katz[11], Johnson and Co-Authors[12], and in a monograph by O'Kelley[13].

Table I lists three methods by which alpha sources may be accurately standardized. Each of these will be described in detail in the following sections. Chemical analysis and calorimetry can, in principle, also be used to determine the amount of an alpha emitting isotope in a source when the isotopic composition and decay constants are accurately known. These methods are limited to sources containing rather large amounts of material, altnough calorimetry, particularly, is a very accurate and precise method for direct measurement of the alpha emission rate of a source. A calorimeter measures the total disintegration energy of the sample if all of the radiation is absorbed in the calorimeter. Alpha rays and the nuclear recoil energy are always completely absorbed and beta rays are also completely absorbed in most calorimeters used for alpha and beta sources. A correction is easily made for energy lost by gamma emission. Good reviews of the calorimetry of radioactive sources have been published by Eichelberger[14] and Gunn[15].

TABLE I

METHODS USED FOR ABSOLUTE ALPHA COUNTING

Method	Application
Low geometry, defined solid angle counters.	Analysis of mounted solid samples and standardization of sources of any alpha emitter having a sufficiently high specific activity.
Alpha-gamma coincidence counting.	Standardization of ^{241}Am alpha sources.
Liquid scintillation.	Analysis of liquid samples for any alpha emitter.

Neither of the above methods can be used to directly calibrate sources of the size used for counting, but rather, are used to prepare a standard solution from which measured aliquots are taken to prepare sources. That procedure is not recommended for preparation of calibration sources because the source mounting step introduces more error than desired. The most sensitive microcalorimeters described in the literature can measure the disintegration rates of sources as small as about 0.1 milliwatt with a precision in the tenths of a percent. Assuming that a 0.1 milliwatt source is needed, the required amount of a typical

alpha emitter such as ^{241}Am would be 3 millicuries or 6.66 x 10^9 d/m. This is at least a factor of 10 greater than the source size desired even for low geometry counting.

2. Low Geometry, Defined Solid Angle Counters

a. Low Geometry Attachments to Proportional and Scintillation Counters

The traditional absolute alpha counter is a defined solid angle counter of low geometry, a device in which the source is positioned a fixed distance from a collimating aperture through which the alpha particles must pass to be counted. There are several variations in physical details and in the type of alpha detector used but the basic counter is a two-chambered device with the collimating aperture separating the chambers. The alpha counting efficiency of such a counter is determined by the solid angle subtended by the collimator and the source with corrections for the area of the source. The accuracy of the calculated geometry factor depends on the accuracy of the measured values, namely, the diameter of the collimating aperture, the source to aperture distance, and the area of the source. Care is taken to avoid errors due to non-uniformity of the deposit on the source, scattering of alpha particles from the sides of the source chamber, and self-absorption of alpha particles in the source.

The use of a low-geometry counter greatly reduces the effect of two sources of uncertainty in the efficiency of counting by 2π and other high geometry counters. These are self-absorption and backscattering. Self-absorption losses are minimized by using sufficiently thin sources and by the geometrical arrangement by which only particles emitted at nearly right angles to the source plane are counted. Backscattered alpha particles are not counted because alpha particle scattering is angle dependent, with virtually no scattering at angles above about 30 degrees to the source plane.

The earlier low geometry alpha counters were made by attaching a vacuum source chamber to a standard proportional counter, such as the Simpson methane proportional counter. Jaffey[11] reviewed this subject in about 1953 and mentioned several kinds of low geometry attachments. He emphasized the importance of an accurately machined metal chamber with a very reproducible source-aperture spacing. Early chambers of this kind are described by Fleming and co-workers[16] and by Curtis and co-workers[17,18]. Fleming, Ghiorso, and Cunningham[16] used their low geometry counter to measure the specific activities of ^{234}U and ^{236}U. Their counter consisted of an evacuated metal sample

chamber attached to a conventional argon-filled alpha counter, with an aluminized mica window covering the collimator at the top of the vacuum chamber. The geometry, calculated to an accuracy of about 0.1 percent from the physical measurements, was about 1/800.

The review on absolute alpha counting by Curtis and co-workers[18] contains good discussions of the limitations of 2π counting, estimates of the backscatter and self-absorption effects, and a description of a low geometry attachment for a proportional alpha counter. In many respects, the counter was similar to that described by Fleming and co-workers[16], as well as those used routinely at Berkeley (Lawrence Radiation Laboratory, University of California), Hanford (AEC plutonium plant at Richland, Washington), and Savannah River (AEC plutonium plant at Aiken, S.C.). It had a parallel plane source-aperture configuration in a cylindrical chamber. The chamber described by Curtis and co-workers differed, however, in having a multiple orifice plate and both a high and a low pedestal source holder to give geometry flexibility. The orifice plate contained four accurately machined holes in a Y pattern with provision for covering the outer three holes when desired. Thus, four geometries were possible with the combination of orifice plate and two source pedestals. For the particular chamber described, the geometry factors (1/G) were: $4.1376 \times 10^2 \pm 0.08\%$, $4.0258 \times 10^3 \pm 0.10\%$, $1.5836 \times 10^3 \pm 0.06\%$, and $1.6022 \times 10^4 \pm 0.10\%$. The geometry values were calculated for each orifice by using formulas similar to those described by Jaffey[19].

The source chamber for a typical low geometry alpha counter is shown in Figure 1. A counter of this design was used by Overman at the Savannah River Laboratory for a recent evaluation of low geometry alpha counting[20] and similar ocunters have been used at Berkeley and Hanford. The chamber had a geometry of about 1/700 and was attached to a methane-flow alpha proportional counter.

H. P. Robinson of the University of California at Berkeley described[21] the precision low geometry counter shown in Figure 2. The source chamber of this counter was 39-1/2 inches long, the collimator had a 3-inch diameter and the geometry was about 1/2600. The chamber had eight concentric baffles to stop alpha particles scattered from the walls. Without baffles, the error introduced by wall scattering would be nearly one percent. The error would probably be much smaller in the counter shown in Figure 1 because of the shorter tube (larger scattering angle) and the small (1/2-inch) collimator used.

Robinson also discussed a medium geometry counter with a geometry of 1/29 having a 4-inch collimator and a 5.1-inch spacing. The use of

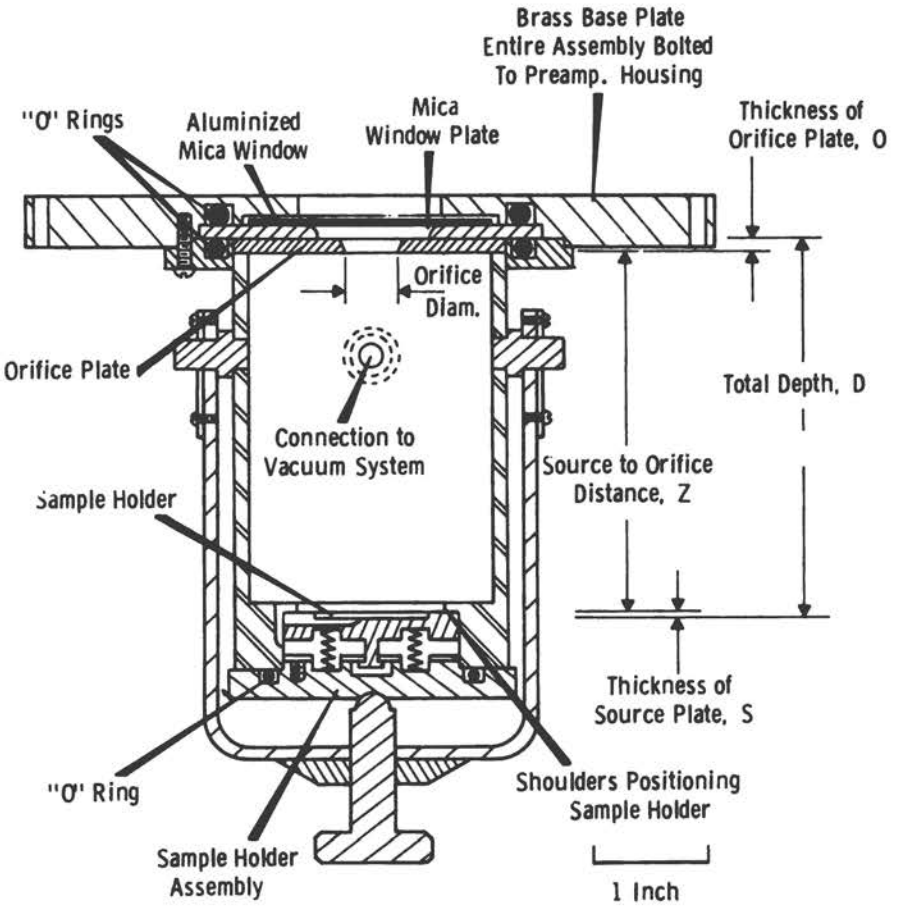


Figure 1
Vacuum Chamber Attachment
(Courtesy of R. F. Overman, Savannah River Laboratory)

such a large collimator area for a given geometry permits a relatively large source-collimator spacing, minimizing the source area correction, since, as the spacing increases, a spread source more closely approximates a point source.

Robinson recommended use of gauge blocks and a microscope for accurate measurement of the chamber dimensions. A circular gauge a few thousandths of an inch smaller than the collimator is made and accurately calibrated using gauge blocks. It is placed in the collimator and the gap measured with a microscope.

Most of the low geometry standard alpha counters described in the literature have had proportional counters attached to the low geometry source chambers. High quality alpha scintillation counters

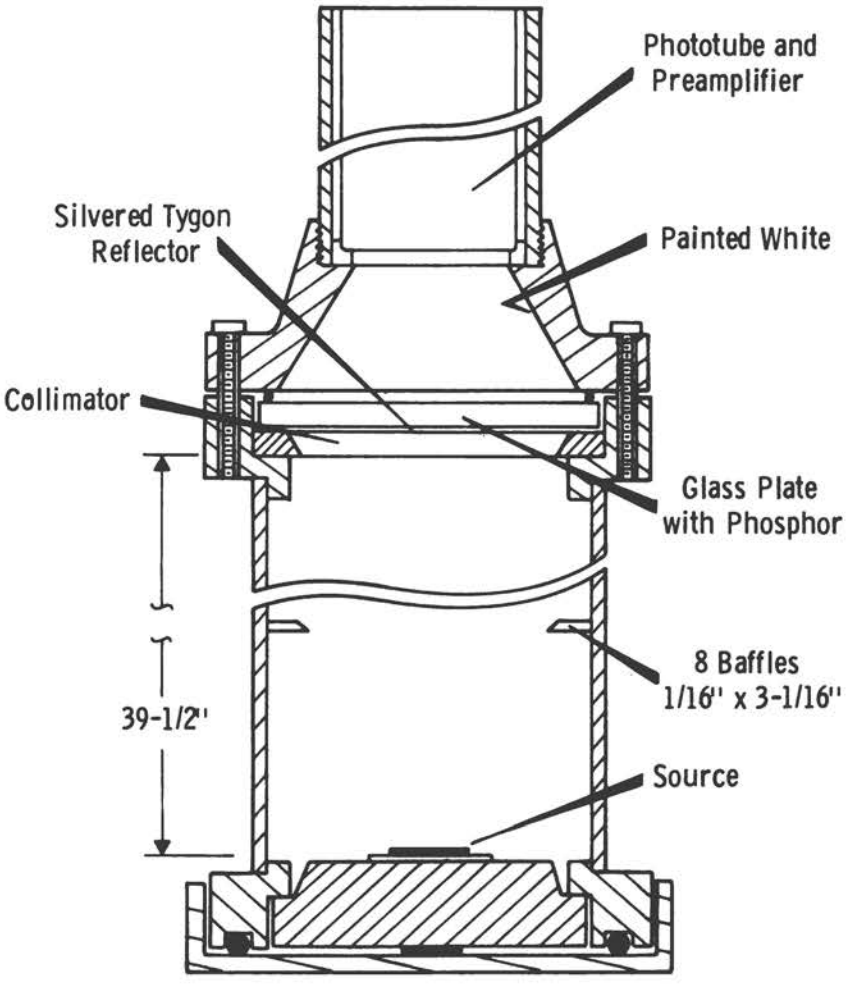


Figure 2.
Low Geometry Scintillation Counter
(Ref. 21)

(especially silver activated ZnS screens) and the necessary high quality multiplier phototubes and electronic accessories are now commercially available and offer some advantages over proportional counters. Therefore, some recently constructed low geometry counters use alpha scintillation counting instead of proportional counting.

The counters described by Robinson[21] had ZnS scintillation screens that were made in his own laboratory (see Fig. 2). The ZnS screen, formed on a glass plate and covered with a reflective film, was placed just above the collimator. The scintillation screen and

reflector film formed the air-tight top of the low geometry, evacuated chamber.

One of the compelling reasons for using a scintillation counter on Robinson's low geometry chamber was that the collimators were large (3 or 4 inches) so that forming a thin vacuum-tight window (e.g., using one or two mg/cm^2 mica) over such a large aperture was probably impossible.

A plastic scintillation screen may be used as an alpha detector instead of zinc sulfide. Thin plastic scintillation screens with uniform response are simple to prepare, whereas good zinc sulfide screens that are uniform and certain of a 100 percent counting yield are difficult to prepare. Plastic scintillators, on the other hand, do not permit good discrimination between α and β particles. Spernol and Lerch[22] described a low geometry absolute alpha counter using a NE 102A plastic scintillator cemented to a glass plate and light-coupled to a multiplier phototube. They tested thicknesses of plastic from 0.01 to 2 mm and carried out experiments with several low geometry vacuum chamber attachments with geometries ranging from 1/50 to 1/30,000. Using a counter with a geometry of about 1/2000 (64 cm source to aperture distance and 6 cm aperture diameter), they showed that alpha sources could be counted with an accuracy of better than 0.2 percent. Average results for four ^{241}Am sources counted by low geometry, 4π alpha, 4π α-γ coincidence, and liquid scintillation methods were shown to agree within the 0.2 percent figure.

b. The Ward Counter

In the Ward counter, the window between the source region and the counting region is omitted and the entire counter is filled with the counting gas (methane) at a low pressure. The pressure of the filling gas is adjusted so the alpha particles come to rest in the counting region. A counter of this kind described in a report by Cruikshank and co-workers[23] had a defining aperture diameter of 0.75 in. and a source distance of 4.877 in. and was operated with methane at a pressure of about 3 cm. The calculated geometry was 1/693.

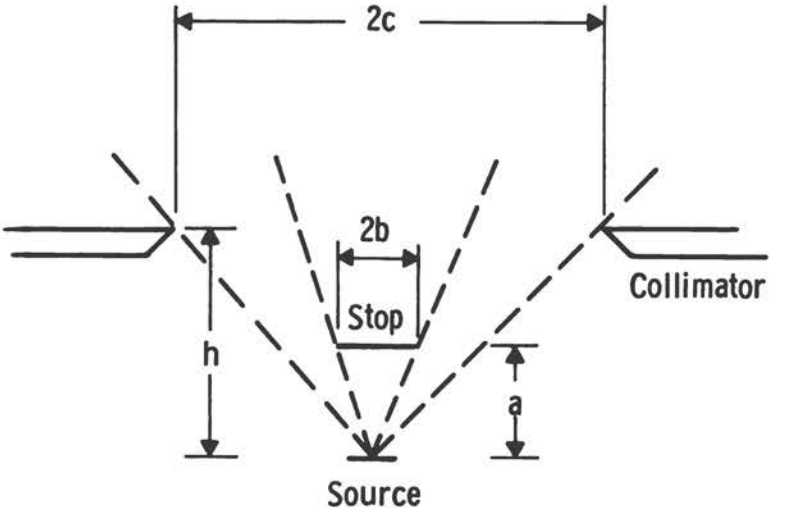
A structurally improved version of the Ward counter is described by Hurst and Hall[24]. The counter had a geometry of 1/(717.3 ± 0.8) for a uniformly spread source of 0.4-inch diameter and was operated with methane at 6 cm. The simplicity of the Ward counter is attractive. The operation of a proportional counter in this manner

is also favorable, requiring low voltage and being capable of adjustment (by pressure setting) to a relatively high pulse height for alphas from the source relative to background alpha and beta activities arising from impurities in the walls of the source chamber.

Cruikshank and co-workers[23] indicated that their Ward counter was calibrated to an accuracy of about ± 0.35 percent. Hurst and Hall[24] discussed using a Ward counter that was accurate to about ± 0.5 percent. They considered the Harwell model of the Ward counter to be accurate to ± 0.1 percent. The high voltage plateaus were very flat and standard sources recounted over a period of more than a year gave identical results.

3. Medium Geometry, Defined Solid Angle Alpha Counters

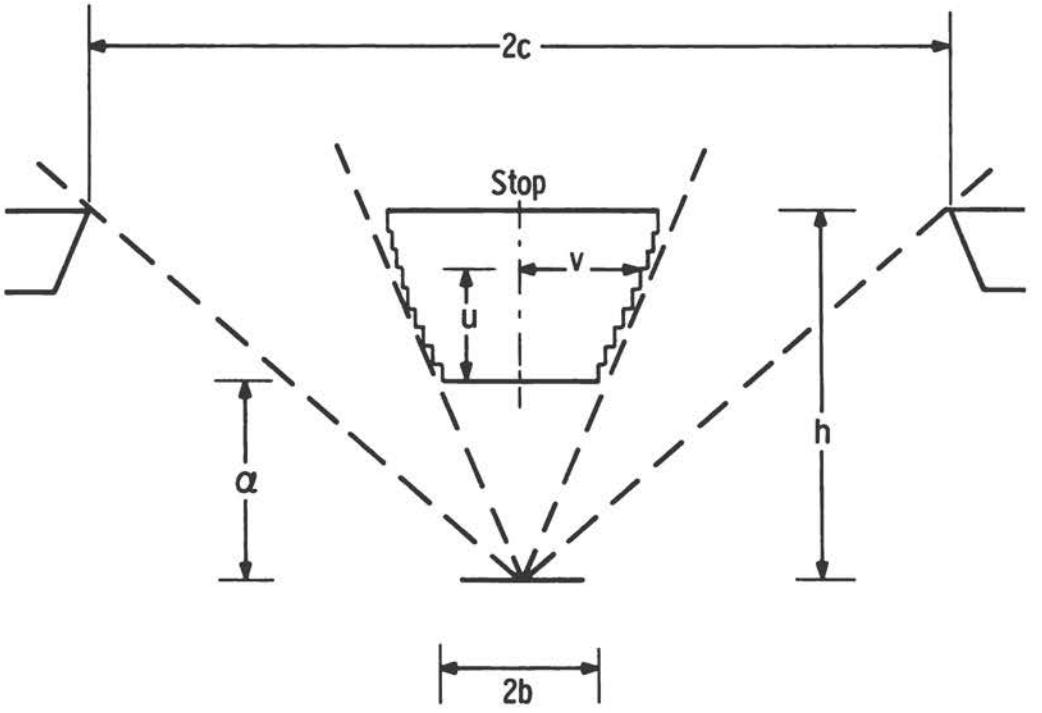
H. P. Robinson[25] designed a relatively high geometry alpha chamber capable of precise absolute counting. The solid angle is



$$a = \frac{b^{2/3}}{c^{2/3}} \sqrt{c^2 + h^2 - c^{4/3} b^{2/3}}$$

Figure 3.
Plan of High Geometry
Defined Solid Angle Counter by Robinson
(Ref. 25)

defined by the combination of a collimator and a central axial stop of such a size and shape and at such a position that the effect of



$$\frac{(u-k)^2}{a^2} + \frac{v^2}{m^2} = 1$$

$$\frac{v^2}{b^2} = -\frac{b^2}{m^2-b^2} \cdot \frac{u^2}{\alpha^2} + 2\,\frac{u}{\alpha} + 1$$

Figure 4.
Solid Stop Design
(Ref. 25)

the position of the source on the solid angle geometry is minimized. The principle of the design is illustrated schematically in Figure 3. The compensating stop minimizes variations in counting geometry due to vertical displacement of the source and counting of backscattered alphas is negligible because the smallest angle subtended by the aperture is above 30°. The design illustrated in Figure 4 shows a solid stop designed to minimize errors due to both horizontal and vertical displacements of the source. The stop has an elliptical

shape with v being the radius at a distance u from the bottom; k, a^2 and m^2 in the equations are arbitrary constants. Robinson constructed a counter with an elliptical stop having a 21.77 mm diameter at the bottom end, a collimator diameter, 2c, of 152.405 mm, a source to stop distance, a, of 21.37 mm and a collimator to bottom of stop distance of 22.17 mm. The counter had a geometry of 0.19748. When in use the source chamber was attached to a ZnS scintillation counter and evacuated. A comparison with a low geometry standard counter gave excellent agreement with the calculated geometry.

4. Calibration of Low Geometry Counters

A simplified formula that may be used to calculate the counter geometry factor for a uniform circular source with radius, s, at a distance, h, from a circular aperture having a radius, r, is:

$$\text{Geometry} = \frac{r^2}{4h^2} - \frac{3r^2}{16h^4}(r^2 + s^2) + \frac{r^2}{h^6}\left(\frac{2r^4 + 3s^2r^2 + 2s^4}{12.8}\right)$$

An exact formula for calculating the geometry for a spread source in such a counter is very complex and many simplified formulas have been derived. The derivation of several of these formulas has been described by Jaffey[19].

Required dimensional accuracies for a geometry accuracy of 0.1 percent are given in Table II. To obtain an accuracy of

TABLE II

DIMENSIONAL ACCURACY REQUIRED FOR A PHYSICAL GEOMETRY ACCURACY OF ± 0.1%, LOW GEOMETRY ATTACHMENT

	Maximum Allowable Error		
Critical Dimension	Relative Percent	Absolute Inches	For Nominal Dimensions (inches)
Aperture (r, radius)	± 0.05%	± 0.0001	0.2000
Height (h)	± 0.05%	± 0.002	4.000
Sample radius (s)	± 8	± 0.03	0.375

± 0.1 percent in the geometry of a counter having a geometry of about 1/700, the aperture radius and height of the chamber must each be measured with an accuracy of about ± 0.05 percent. That is difficult to achieve. In addition, some uncertainty in the effective aperture diameter may occur because alpha particles will penetrate up to about

10 microns of steel and be counted. The knife edge of the aperture should therefore have as high an angle as possible. An additional source of error stems from uncertainty about the amount of low angle alpha particle scatter from the walls of the chamber. A series of circular baffles should be used to reduce the error.

The reproducibility of dimensional measurements of low geometry chambers at Hanford has indicated that their calibrations are accurate to within about ± 0.3 percent[26].

5. Alpha-Gamma Coincidence Counting

a. Principle

Coincidence counting has been used to determine the absolute disintegration rates of beta radioactive sources for several decades. Electronic coincidence methods date from about 1930. Geiger and Werner used a visual coincidence counting method for alpha scintillations as early as 1924, as mentioned previously in the Introduction. Descriptions of the theory and practice of coincidence counting are found in numerous books and review papers. The principle of the coincidence method of calibration can be illustrated most easily with the simple use of a beta emission followed by a single gamma ray. In the case of absolute beta counting, a beta detector, a gamma detector, and the source are arranged in such a way that both the beta and gamma counting rates are high enough for good counting statistics and low background corrections. The observed net beta count rate, R_β, is:

$$R_\beta = A_o \varepsilon_\beta$$

where A_o is the absolute disintegration rate and ε_β is the efficiency of the beta counter. Similarly, the net gamma count rate, R_γ, is:

$$R\gamma = A_o \varepsilon_\gamma$$

and the coincidence counting rate is:

$$R_c = A_o \varepsilon_\beta \varepsilon_\gamma$$

Solving these three equations in terms of the measured beta, gamma, and coincidence counting rates, the disintegration rate of the sample is given by:

$$A_o = \frac{R_\beta R_\gamma}{Rc}$$

The observed beta, gamma, and coincidence counting rates must be corrected for background, chance-coincidence count rates, beta counts,

if any, detected by the gamma counter, and gamma counts detected by the beta counter. These corrections can be minimized by judicious choice of source intensity and counter characteristics. The chance coincidence count rate is given by:

$$C = 2T\, R_\beta R_\gamma$$

Where T is the coincidence resolving time of the counter.

When the coincidence count method is applied to absolute alpha counting, or actually, alpha source standardization, ^{241}Am is used as the alpha source. The decay scheme of ^{241}Am is shown in Figure 5[27]. The alpha particle spectrum of ^{241}Am has several alpha energy levels corresponding to excited states of ^{237}Np. The most abundant gamma ray of ^{241}Am is at 60 keV, with less abundant rays at 27, 33, 76, 103, and others. The alpha-gamma coincidence counting method is usually applied to the 60 keV gamma, only, the other gamma rays being excluded by energy selection. The abundance of the 60 keV gamma is 0.36[27].

b. Application

The first reported application of α-γ coincidence counting to calibrate ^{241}Am alpha sources was by Lyon and Reynolds[28]. They used a 4π alpha proportional counter mounted above a sodium iodide gamma detector to count ^{241}Am sources mounted on thin plastic films. The $4\pi\alpha$ counter was also used in a $2\pi\alpha$ counter mode and identical results were obtained in standardizations made using $4\pi\alpha$-γ coincidence and $2\pi\alpha$-γ coincidence counting. Corrections were necessary for source mount interferences in the $4\pi\alpha$ counter. Calibrations by $2\pi\alpha$-γ coincidence of sources mounted on platinum and stainless steel disks were also made.

Use of the α-γ coincidence method was reported by Overman[20], by Sakai and co-workers[29], and by several laboratories that participated in an international exchange of standard alpha sources of ^{241}Am, as reported by Rytz[30]. Overman used both a 2π windowless methane flow alpha counter and a ZnS alpha scintillation counter in combination with a NaI gamma scintillation counter for his source calibrations. The results agreed within about 0.5 percent with calibrations made by low geometry alpha counting of the same sources. His data are shown in Table III.

Sakai and co-workers[29] used silicon p-n junction-type detectors for the alpha counting and NaI scintillation counters for the gamma counting. The purpose of their work was to measure the half-life of

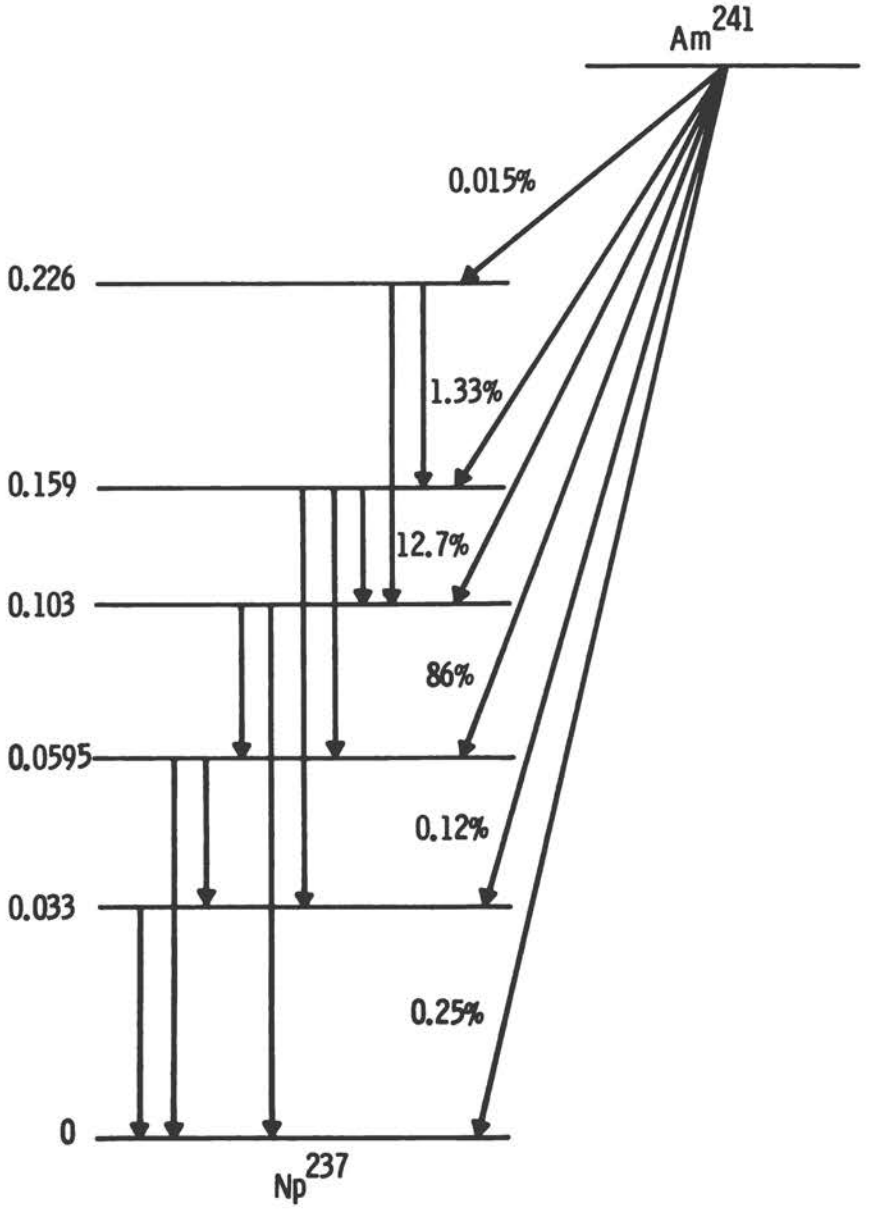


Figure 5
Simplified Decay Scheme of ^{241}Am

the 60 keV level of ^{237}Np and to measure the effect of the coincidence resolving time on the absolute measurement of ^{241}Am source activities. No exact comparisons with other methods of source calibration were made although some comparative measurements were made with a low geometry counter arrangement of limited accuracy.

TABLE III

COMPARISON OF COUNTERS
WITH ^{241}Am SOURCES*

(Reference: DP-780, R. F. Overman)[20]

Source	Coincidence Counting d/m		Low Geometry Counting d/m
	Counter I	Counter II	
1.	$1.1068(10^5)$	$1.1053(10^5)$	$1.1033(10^5)$
	± 0.36%	± 0.07%	± 0.05%
2.	$1.1895(10^5)$	$1.1894(10^5)$	$1.1809(10^5)$
	± 0.49%	± 0.36%	± 0.04%
3.	$1.1514(10^5)$	$1.1566(10^5)$	$1.1489(10^5)$
	± 0.66%	± 0.37%	± 0.15%
Differences**	+ 0.46%	+ 0.62%	--

*The precision values are based on the standard error of the mean count rates.

**Weighted average of coincidence counting results minus weighted average of low geometry counter results.

c. Calculations

Calculation of the absolute alpha disintegration rates of ^{241}Am sources is made by the usual formula:

$$A_o = \frac{R_\alpha R_\gamma}{R_c}$$

which is derived as follows:

Let D_1, D_2, D_3, ... be the disintegration rates of the α particle groups going to the 60, 103, 159, ... keV levels. Therefore the observed alpha counting rate:

$$R_\alpha = D_1e_1 + D_2e_2 + D_3e_3 + ...$$

where e_1, e_2, e_3, . . . are the alpha detection efficiencies. Since the alpha energies are very close to each other, e_1, e_2, e_3 . . . . . have the same values, and

$$R_\alpha = e_1(D_1 + D_2 + D_3 + \ldots)$$

Let E_1, E_2, E_3, ... be the emission rates of the gamma rays at 60, 103, 159 . . . keV; then the gamma counting rate is:

$$R_\gamma = E_1f_1 + E_2f_2 + E_3f_3 +$$

where f_1, f_2, f_3, ... are the gamma ray detection efficiencies. If the efficiencies f_2, f_3, . . are made equal to zero by energy discrimination:

$$R_\gamma = E_1f_1$$

The coincidence counting rate is expressed by:

$$R_{\alpha\gamma} = E_1e_1f_1$$

This equation contains E_1 rather than D_1 because the former rate is the smaller of the two (E_1 is 0.36 while D_1 is 0.86). Since E_2, E_3, ... are rejected by energy discrimination, these terms drop out of the coincidence count expression. Consequently, the three measured values, R_α, R_γ, and $R_{\alpha\gamma}$ can be used to solve the equation.

$$\frac{R_\alpha R_\gamma}{R_{\alpha\gamma}} = \frac{e_1(D_1 + D_2 + D_3 + \ldots\ldots)(E_1f_1)}{E_1e_1f_1}$$

$$= D_1 + D_2 + D_3 \ldots\ldots$$

and this sum of all alpha disintegration rates is equal to the total alpha disintegration rate.

Each measured alpha and gamma count rate (R_α and R_γ) must be corrected for background counts in the usual manner. In addition, a correction must be applied to $R_{\alpha\gamma}$ for chance coincidences. This is calculated from the α and γ count rates and the resolving time, T, of the coincidence counting system

$$C_{ch} = 2T\, R_\alpha R_\gamma$$

where R_α and R_γ are uncorrected count rates.

6. Liquid Scintillation Counting

Several organic liquid scintillators as solutes in liquid organic solvents are used for 4π scintillation counting. Typical liquid scintillators are p-terphenyl, 2,3-diphenyloxazole (DPO or PPO) and 2-phenyl-5-biphenyloxazole at concentrations of 4 to 8 g/ℓ in toluene, xylene, dioxane, or other solvents. A small amount of an

organic compound that serves as a wavelength shifter, such as diphenylhexatrene or 1,4-di-[2-(5-phenyloxazolyl)]-benzene (POPOP) is often added to the scintillator. The excitation produced in the solvent by the nuclear radiation is transferred to the solute before quenching can occur. The solute then loses the excitation energy by producing photons at a wavelength characteristic of the solute. The shifter increases the wavelength to a region nearer the peak of the sensitivity curve of the multiplier phototube. Much more complete treatments of liquid scintillation counting are available in several books and reviews[31,32].

Liquid organic scintillators are valuable for counting weak beta and alpha particles because the problems of self-absorption in sample mounts and absorption in counting tube windows is eliminated. The liquid scintillation counter has considerable versatility although it produces smaller pulse heights than most crystals and solid organic scintillators. Many organic and aqueous solutions can be dissolved directly in the scintillator without serious quenching.

Basson and Steyn proposed in 1954[33] that a liquid scintillation counter be used for absolute alpha counting of solutions. They measured the pulse height distribution for 5.3 MeV (^{210}Po) alpha particles in a scintillator of 2,5-diphenyloxazole in p-dioxane (3 g/ℓ) at room temperature and showed that the absolute alpha count rate could be obtained by extrapolating the integral count rate to zero bias. The alpha plateau was not flat enough for direct absolute counting. In 1956, Basson published data on the absolute 4π alpha scintillation counting of astatine-211 solutions[34]. In that work, he obtained very flat plateaus and a complete separation of the two principal alpha peaks of ^{211}At from background. Both terphenyl in toluene and 2,5-diphenyloxazole in p-dioxane were used.

Horrocks and Studier[35] applied the liquid scintillation counting method to the determination of ^{241}Pu in plutonium samples by means of the alpha count-beta count ratio. They used 4 g/ℓ of p-terphenyl in xylene with 0.1 g/ℓ of POPOP as the scintillator and operated at -9°C to reduce the background count rate. In addition, they used a coincidence arrangement to improve the counting efficiency for the weak beta particles of ^{241}Pu and further reduce background. These refinements would not normally be required for alpha counting alone. The alpha peak of the plutonium isotopes (^{239}Pu, ^{240}Pu, and ^{242}Pu) was completely separated from the beta spectrum and the authors estimated that only about 0.7 percent of the α-particles counted in the ^{241}Pu beta energy region.

Seliger[36] measured the energy resolution for alpha and beta particles and demonstrated 100 percent alpha counting in the liquid scintillator consisting of phenyl-biphenyloxadiazole-1:3:4 and POPOP in toluene (8 g/ℓ and 0.1 g/ℓ, respectively). He obtained a conversion efficiency of one photon per 125 eV of beta and one photon per 1400 eV of alpha energy. After selection of conditions that gave a flat alpha plateau, agreement between liquid scintillation counting and 4π proportional counting of ^{210}Po solutions was within 0.3 percent.

Ihle and co-workers[37] also compared the results of absolute alpha counting by 4π liquid scintillation counting and 4π proportional counting, obtaining agreement within a few tenths percent or better in all cases.

Brauer, Stromatt, Ludwick, et al[38,39] used the liquid scintillation method to measure the specific alpha activity of ^{237}Np. An apparatus similar to that of Horrocks[35] was used and the scintillator solution was 75% dioxane, 12.5% anisole, 12.5% 1,2-dimethoxyethane, 7 g/ℓ PPO and 50 mg/ℓ POPOP. The specific alpha activity obtained agreed with that obtained by standard low geometry alpha counting within the counting precision at the 95 percent confidence level.

An important limitation of liquid scintillation counting is that an alpha particle produces a pulse size about $1/11^{th}$ of that produced by a beta particle of the same energy. That, coupled with the rather poor energy resolution of liquid scintillators, results in the possibility of serious beta interference when mixed alpha-beta emitters are to be counted. In all applications, a careful check of the alpha plateau must be made with all impurities to be added with the sample present in the scintillator.

7. Intercomparison of Alpha Standardizations

The following tables of data illustrate typical agreement between different methods of alpha standardization by various laboratories.

Table IV

Intercomparison of Four Low Geometry Ward Chambers at Chalk River[40]

Counter	^{241}Am Source
1	5.068×10^6 d/m
2	5.063×10^6 d/m
3	5.059×10^6 d/m
4	5.075×10^6 d/m

Table V

Intercomparison of Three Alpha Sources Between Two Laboratories

(from Glover and Hall)[41]

Source		Harwell Values* d/m	Berkeley Values** d/m
S-36		1.956×10^6	1.956×10^6
S-37	(1)	1.169×10^6	1.164×10^6
	(2)	1.169×10^6	
S-49	(1)	8.109×10^6	8.113×10^6
	(2)	8.110×10^6	

*Ward type counters. Two counters used for S-37 and S-49.

**Lawrence Radiation Lab.; low geometry scintillation counters used.

TABLE VI

Intercomparison of Results by Several Laboratories Using Three Plutonium Sources Prepared at Aldermaston (Low Geometry Chambers Used)

(Reference: AWRE-O-59/63)[42]

Laboratory	Average Count Rate, Relative to AWRE Value
AWRE	1.000
AECL (Chalk River)	1.014
AERE (Harwell)	0.999
DERE (Dounrey)	1.001

TABLE VII

Comparison of Alpha Sources
(Reference: HW-59642)[38]

Source	Hanford	Berkeley*	Argonne National Laboratory**
3 (^{239}Pu)	4.107×10^5		4.128×10^5
4 (^{239}Pu)	4.216×10^4		4.238×10^4
10 (^{241}Am)	4.705×10^6	4.713×10^6	
12 (^{241}Am)	3.731×10^7	3.728×10^7	

*H. P. Robinson, private communication.
**R. F. Buchanan, private communication.

TABLE VIII

INTERCOMPARISON OF METHODS ON ^{237}Np SOURCES

(Reference 39)

Measured Specific Activity of ^{237}Np Source
(d/m/μg)

Low Geometry Proportional Counting	Liquid Scintillation Counting
1591	1583
1586	1575
1574	
1587	
Mean 1584	

TABLE IX

Intercomparison of Standardizations of a ^{241}Am Solution (Note 1)
(Reference: NP-14872)[30]

	d/s/mg
A. Values obtained by defined solid angle alpha counting.	11.3[2] 11.21
B. Values obtained by liquid scintillation counting	11.3 11.57 11.29 11.28 11.12[2] 11.27 11.22[2]
C. Values obtained by $4\pi\alpha$-γ coincidence counting.	11.31 11.28 11.28 11.30[2] 11.22[2] 11.32 11.36 11.30 11.45 11.23 11.32 11.27 11.31 11.26 11.26 11.27 11.27

Note 1: 21 laboratories throughout the world participated in this exchange sponsored by

the International Bureau of Weights and Measures. For further details and identification of the laboratory that submitted each result, see the original paper. Each value is the result reported by one laboratory.

Note 2: Estimated relative error greater than 0.5 percent.

D. PREPARATION OF ALPHA CALIBRATION SOURCES

Calibrated sources of alpha active material are needed to standardize the alpha counters used in routine work. The absolute alpha disintegration rates of the sources are measured by one of the absolute counting methods described in the foregoing sections. Because alpha particles have a short range in matter, the source must be very thin and uniformly deposited. In addition, the alpha mount backing must be flat and smooth to avoid absorption losses when used on high geometry counters since scratches, indentations, and ridges block alpha particles emitted at low angles.

Calibration sources are most commonly prepared on metal foils although glass and plastic foils may be used. To minimize corrections for alpha backscatter differences, the calibration foils should be of the same material as used for routine source mounting. The calibration foil should be thick enough to be rigid and not susceptible to bending or dimpling. Ten-mil thicknesses are generally adequate. If the same foil thickness is not used for sample mountings, a calculated correction may be needed to correct for the differences in source to counter window distance. The difference is negligible in the case of 2π windowless counters, such as proportional counters, but with a scintillation counter operating in the 30-40 percent geometry range, a difference of 5 mils disk thickness will cause a geometry error of several tenths percent.

A mirror-bright finish on metal foils is necessary and the foil preparation process must not cause visible etching. A disk commonly used at Hanford for routine alpha mounts is a 22 mm diameter stainless steel disk punched from 10 to 12-mil-thick polished sheet with a high mirror surface finish (a No. 4 microinch finish is satisfactory). Platinum is also a favorite foil material since most aqueous and organic solutions can be evaporated on it without causing corrosion, and it can be ignited to red heat in air to remove organic deposits. Platinum sheet is often not smooth enough as purchased for high quality alpha counting foils. The disks may be polished by hand with

fine abrasives. A vibratory metallographic polisher is convenient and a good series of abrasives is No. 600 emery, No. 000 emery, 6 micron diamond (AB Metadi), and MgO powder, used in that sequence.

The deposit of alpha-active material on a source must be uniform and thin to avoid serious loss of alpha particle energy in the deposit. In 2π counting, the tolerable amount of solids on the source is limited to about 30 $\mu g/cm^2$. With a low geometry counter, considerably greater thickness can be accepted because the low angle alpha particles that would be most affected by the source thickness are not counted. A source thickness of up to a few mg/cm^2 can be tolerated in that case (typical ranges of alpha particles of 5 MeV energy are about 8 mg/cm^2 for Al and about 18 mg/cm^2 for Pb) but such a source is not suitable for standardizing 2π counters. Long-lived alpha emitters like ^{238}U, ^{235}U, and ^{232}Th do not make suitable alpha sources for calibration by low geometry counting because even the maximum allowable amount of material per source would yield a rather low counting rate in low geometry counting.

Considerable effort has gone into development of techniques of preparing thin, uniform films of material, especially by scientists making targets and foils for measurement with particle accelerators and reactors.

A good series of summary papers on the methods in common use is available in the proceedings of a siminar held at Harwell in October, 1965[43]. Vacuum evaporation and electrodeposition are widely preferred methods. The former gives the most uniform deposits while the latter gives higher yields, conserving scarce isotopes, and requiring simple apparatus that is readily available in all laboratories. Descriptions of these and a few other good methods for alpha and fission counting source preparation follow.

1. Source Mounting by Evaporation

The most straightforward approach to preparation of alpha sources is evaporation of an aliquot of solution on a suitable backing. However, producing a thin source by that technique is very difficult. First, the solution must be essentially free of salts that would contribute appreciable mass to the final deposit, and second, any solids present, including the active material itself, do not deposit uniformly but tend to deposit as crystals and aggregates that cause self absorption of alpha activity. Spreading agents can be added to the solution during evaporative deposition to reduce the

crystallization problem[44,45]. Tetraethylene glycol or a solution of Zapon* lacquer added to the foil with the active solution inhibits crystal growth and causes thickening to a viscous polymer before the evaporation is complete. The technique results in a deposit containing much organic material which must be burned off. As a result, the deposits tend to be poorly adherent to the backing and require a thin coating of collodion to prevent loss of activity during use.

Glover and Borrell[46] obtained quite adherent coatings of uranium and plutonium by applying many thin layers using cellulose nitrate as a spreading agent. They painted on layers of the active mixture, ignited them, and rubbed the surface with a tissue between successive layers. The deposits had very good uniformity by an autoradiography test.

Direct evaporation of organic solutions can give nearly solid free deposits of some alpha emitters. Examples are ether solutions of uranium and benzene or toluene solutions of plutonium-TTA chelate. Such solutions have a tendency to spread and go over the edges of the foil when heated, rather than remain in drops like aqueous solutions. Placing a common iron washer under the disk on a hot plate so the heat is conducted mainly to the outer rim of the disk helps reduce spreading. A special electrical heating device that heats only the outer rim while a cold spot or heat sink is in contact with the center of the disk is much more effective[47]. In any case, the organic liquid must be added to the disk in small increments during evaporation. Alpha foils prepared by evaporation of organic solutions usually have very good alpha energy resolution and therefore would probably be quite satisfactory as calibration disks.

2. Electrodeposition

The method most widely used by radiochemists for the preparation of standard alpha sources is electrodeposition. The method is applicable to many elements and electrodeposits can be made on foils of noble metals, copper, lead, steel, aluminum, and probably several other metals. Procedures for a few elements were reported in the published records of the Manhattan Project[44,45]. Uranium is electrolytically reduced from the hexavalent uranyl ion to the tetrapositive state at the surface of the mounting foil, which is the cathode. The uranium is deposited as a hydrous oxide or as UF_4, depending on the electrolyte used. The foil is finally heated to convert the deposited uranium compound to the anyhydrous state or to

*Nitrocellulose in acetone.

an oxide. The electrodeposition can be carried out from a 0.06 M ammonium oxalate solution at 80°C and a current density of 120-150 mA/cm^2, a 0.1 N KOH-0.1N K_2CO_3 solution at 5 mA/cm^2, a 0.02 M NaF solution at 5 mA/cm^2, or a saturated LiF solution at 1 mA/cm^2. The procedures that use high current densities are much more rapid than the others. All are reported to give firm, uniform deposits with essentially 100 percent yields.

The early electrodeposition methods for plutonium were based on the same reaction mechanisms as the uranium method. The plutonium was oxidized chemically to Pu(VI) and then electrolytically reduced in a 0.12 M KOH or a 0.2M NH_4OH-NH_4Cl solution at about 5 mA/cm^2. The recovery of plutonium was not complete in these procedures. Miller and Brouns[48] modified the above plutonium electrodeposition procedure and obtained quantitative deposition of the plutonium, using ozone for the oxidation and carrying out the electrolysis in a KOH solution of 1 to 2 N.

Ko[49] developed a general electrodeposition method for actinide elements and applied it to the elements thorium through curium. They are plated as hydrous oxides from a buffered, slightly acid solution without preoxidation. The method relies on a pH change near the cathode due to hydrogen discharge, causing formation of a hydrous oxide of the element on the cathode surface. Many variations of that method have been published[50-57]. The yields are generally less than 100 percent, although Donnan and Dukes[56] developed a carrier procedure that increased the yield to an average of about 99.8 percent. The 30 µg of uranium carrier used in the Donnan-Dukes procedure deposits along with the active material so this method is not recommended for standard sources, although useful in other applications.

Solutions of ammonium formate containing a small amount of free acid were used as electrolytes by Ko[49], the choice of acid, electrolyte acidity, and current density varied from element to element.* A 0.4 M ammonium acetate electrolyte (a pH of about 7) was used for uranium deposition by Brodsky, Fagg and Hanscome[50]. They reported that good deposits were made on aluminum, first coated with zincate solution, as well as on platinum foils.

Most of the other electrodeposition methods for actinide elements also use weakly acidic electrolytes, but Mitchell[52] used a pH of about one and used a very high current density, thereby completing the electrodeposition in 10 to 15 minutes. Khlebnikov and Dergunov[55]

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*See Appendix A.

used an oxalate solution at a pH of 8 to 9 for U, Np, Pu, and Am depositions, and Getoff and Bildstein[57] used isopropyl alcohol solutions for plutonium depositions. Any of these methods will apparently satisfy the primary need of a thin and uniformly deposited source that adheres well to the foil.

The apparatus used for electrodeposition is quite simple. A cell is used in which the metal foil is the cathode with only one side exposed to the electrolyte. The anode is always platinum and it

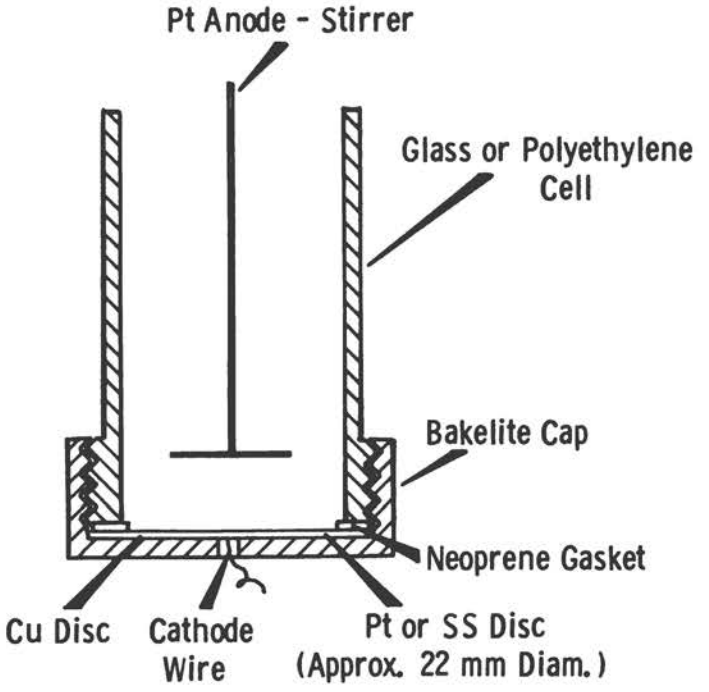

Figure 6
Electrodeposition Cell

generally also serves as the stirrer in the form of a rotating coil or disk. A simple and convenient cell, illustrated in Figure 6, is made by cutting the bottom off a screw-capped glass vial. A plastic vial can be used in the same way. A similar cell, shown in Figure 7, can be made from a Teflon or lucite rod and a screw cap of brass or copper, as illustrated. The latter cell is designed to provide an electrodeposit area that is smaller than the internal cross section area of the bulk of the cell by tapering the interior walls near the bottom. A vertical glass chimney held by 3 or 4 springs or rubber bands against the disk on a flat metal plate is also commonly used. The

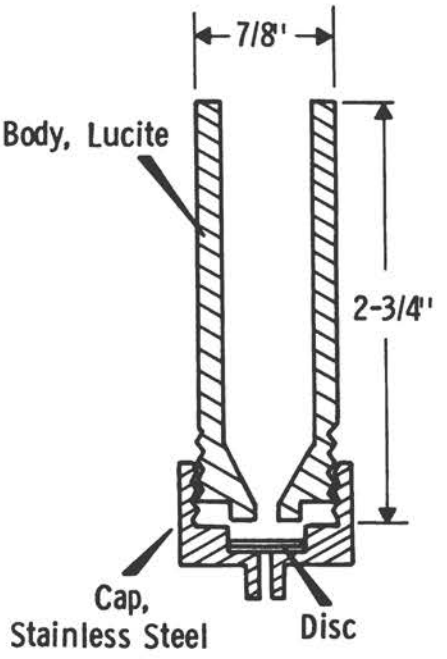

Figure 7
Plastic Electrodeposition Cell

bottom rim of the chimney is first coated with rubber cement to form a gasket. The electrolyzing current may be supplied by a simple dc power source with a rheostat, a milliameter (about 0-200 mA range) and a voltmeter in the circuit. A 12-volt battery is sufficient for most aqueous electrolyte procedures, although high current density procedures like that used by Mitchell[52] require more voltage. Commercial electroanalyzer systems are also convenient sources of power.

3. Sublimation

Very uniform sources of most materials can be prepared by subliming them in a vacuum in an arrangement that allows the vaporized material to condense on the desired foil. Any foil material, including plastic film, can be used and the size and shape of the deposit can be easily varied by using suitable masks. The yields are generally quite low, although Pate and Yaffe[58] and Parker[59] designed crucibles that collimate the molecular beam and give nearly 100 percent yields.

The use of the high temperature sublimation technique was mentioned by Asaro and co-workers in 1952[60] in connection with preparation of very thin foils of americium and curium chlorides for

alpha spectrometry. Milsted[61] described apparatus and evaluated the results of source preparations for elements from thorium to curium. Jackson[62] carried out similar evaluations and also described an improved version of Milsted's sublimation apparatus. Other descriptions of the technique have been published by Salo and Garden[63], Parker[59], and others[43].

The commonly used sublimation method is a high temperature flashing of an inorganic compound of the element desired from a resistance heated filament in a vacuum. Apparatus and techniques vary considerably but the following is a typical procedure. A thin tungsten or tantalum filament shaped to a V-trough is clamped between water cooled electrodes in a vacuum chamber such as a commercial vacuum evaporator. The desired isotope is placed in the filament trough by pipetting a solution or a slurry of an oxide or other compound. The filament is then heated in the vacuum by passing current through it, first to a low red heat to remove organic matter and other volatile impurities, and finally to a high temperature to vaporize the desired isotope. The target foil is either not present or is masked during the first step. The foil is located above the filament in contact with a cooled plate or heat sink.

The foil is generally masked by an aperture plate that determines the area of the deposit. Filament temperatures between 2000 and 3000°C are used and even the actinide oxides vaporize under vacuum in this temperature range. After use, a couple ten-second heating periods at the maximum temperature of the filament thoroughly clean it for reuse.

Crucibles of graphite, ceramics, or metals can be used instead of filaments, with the advantage in some cases of better control of yields. Parker[59] described a crucible designed to collimate the molecular beam and give essentially 100 percent yield. The crucible can be heated to about 1000°C with a resistance winding and then electron bombarded to raise the temperature farther. A similar apparatus is described by Kobisk[43].

The chlorides and oxychlorides of the actinide elements are more volatile than the oxides so dried deposits from chloride solution on the filament are generally preferred. However Milsted[61] mounted nitrates and pre-flashed the filaments at temperatures high enough to remove organic impurities and alkali salts but below the point at which the oxides sublimed.

The isotope to be mounted must be free from appreciable amounts of impurities because all salts mounted on the filament will be deposited on the foil and will increase the source thickness. Final

purification of the isotope is usually carried out by an extraction or ion exchange step. For example, uranium, plutonium, or neptunium can be absorbed on an anion exchange resin, washed with an 8 to 12 M HCl solution and then eluted with 1 to 2 M HCl[64,65]. The transcurium actinides can be absorbed on a cation exchange resin, washed free of extraneous ions with dilute HCl, and desorbed sequentially first with 2 M HCl and finally with 6 M HCl[66].

Certain organic compounds such as the acetylacetonates of uranium and plutonium, and chelates of several elements, can be sublimed at low temperatures in a vacuum[44,58] to form thin deposits on metal or plastic foils. To eliminate the organic material in the deposit and more permanently fix the radioactive material, a chemical treatment such as vapor bromination followed by ignition is necessary.

4. Other Techniques

A unique method called electrospraying can be used to deposit small amounts of an isotope onto a metallic foil or disk. The method is described by Carswell and Milsted[67], Blumberg, Stein and Gursky[68], and other scientists[43]. An organic solution of the isotope is drawn into a very fine capillary and ejected as a fine spray by applying a potential of several thousand volts between the solution and a foil placed a few centimeters below the tip of the capillary. The organic liquid apparently evaporates before the fine droplets strike the foil. The correct choices of voltage and capillary size are made by trial and error. A one mm ID capillary with a 0.2 mm tip is typical.

Adsorption of a nuclide from a dilute solution onto a foil can be used to produce infinitely thin deposits (a few tenths of a monomolecular layer in average thickness). El Guebely and Sikkeland[69] demonstrated the method with ^{239}Pu and ^{233}U in dilute HCl solutions. Glass, aluminum and platinum surfaces were used. The maximum amount of material that could be deposited was of the order of one ng/cm^2.

II. ABSOLUTE MEASUREMENT OF SPONTANEOUS FISSION EVENTS

A. Characteristics of the Spontaneous Fission Process

The possibility of spontaneous fission of heavy elements was forecast by Bohr and Wheeler in 1939[70] and was later observed by Flerov and Petrzhak[71]. The systematics of nuclear decay by fission has been the subject of many papers[72-74]. The stability of nuclides

TABLE X

Typical Spontaneous Fission Half-Lives
(values in years unless otherwise noted)

Isotope	Alpha Half-Life*	Spontaneous Fission Half-Life**
^{232}U	71.7	8×10^{13}
^{234}U	2.48×10^{5}	1.6×10^{16}
^{235}U	7.13×10^{8}	1.8×10^{17}
^{236}U	2.39×10^{7}	2×10^{16}
^{238}U	4.51×10^{9}	5.9×10^{15}
^{236}Pu	2.85	3.5×10^{9}
^{238}Pu	86.4	4.9×10^{10}
^{239}Pu	2.44×10^{4}	5.5×10^{15}
^{240}Pu	6240	1.34×10^{11}
^{242}Pu	3.79×10^{5}	7.06×10^{10}
^{244}Pu	7.6×10^{7}	2.5×10^{10}
^{240}Cm	26.8 days	1.9×10^{6}
^{242}Cm	163 days	7.2×10^{6}
^{244}Cm	18.1	1.4×10^{7}
^{246}Cm	5480	2×10^{7}
^{248}Cm	4.7×10^{5}	4.6×10^{6}
^{249}Bk	314 days	6×10^{8}
^{246}Cf	35.7 hr	2×10^{3}
^{248}Cf	350 days	7×10^{3}
^{250}Cf	13.2	1.5×10^{4}
^{252}Cf	2.65	82
^{254}Cf	-	60.5 days

* Ref. 75.

** Data selected from Ref. 76.

with regard to spontaneous fission decreases with increasing atomic number; thus, most nuclides that have been observed to fission spontaneously are transuranium elements. In general, the logarithm of the spontaneous fission half-life decreases linearly with increasing Z^2/A for even-even nuclides[72]. Decay of odd Z- or A-number nuclides by spontaneous fission is much less common and those that have been measured have much longer half-lives than their neighbors.

Table X lists the spontaneous fission and alpha half-lives of the elements through californium that have measured spontaneous fission

rates. Note that the alpha decay rates are several orders of magnitude greater than the spontaneous fission rates.

The kinetic energies of fission fragments range from about 50 to 100 MeV, in contrast with alpha particles which have energies ranging up to about 10 MeV. The mean kinetic energies of the low and high mass groups of fission fragments are approximately 61 and 93 MeV, respectively. A fission fragment has an initial net charge of about + 20e, but the charge decreases continually during the slowing down process in an absorber. In that respect, the ionizing characteristics of fission particles differ from those of alpha particles. As a consequence, a fission fragment produces a rather uniformly high ion density along its entire path in a gas while an alpha particle produces a lower ion density along most of its path but the density increases sharply to a maximum near the end of the path. The ion density in the first part of the fission particle path is high because of the high charge, as predicted by the Bethe-Livingston equation[10]:

$$\frac{-dE}{dx} = \frac{4\pi e^4 z^2 NB}{mv^2}$$

Where $\frac{-dE}{dx}$ is the energy loss per unit path in a gas, z is the number of units of charge e, N the density of the absorber in atoms/cm^3, and B is a "stopping power" term. Fission particles also lose energy by nuclear collisions. Ionization due to collisions continues along the path, unaffected by charge, but increasing with decreasing velocity, just as the electronic ionization process increases with decreasing velocity, as indicated by the equation above. A good discussion of fission particle ranges and the theory of the energy loss mechanism is given in a paper by Bethe and Ashkin[9]. Since the fission particles have a range in air (at STP) of from 2 to 3 cm and alpha particles have ranges from 4 to 5 cm, ionization chamber dimensions may be selected so that the last half of an alpha particle path, and more than half of its ionization, are cut off without interfering with the fission particle path. That enhances the fission fragment to alpha particle pulse height ratio.

The energy distribution of the fission fragments in spontaneous fission does not differ measurably from that of induced thermal neutron fission of the same nuclide nor apparently from that of induced fission of neighboring nuclides of the same element. That was verified for the case of ^{240}Pu vs ^{239}Pu and ^{242}Pu vs ^{241}Pu (same nuclide is fissioned in these cases)[73,77], and for ^{238}U vs ^{235}U [74].

B. Applications of Spontaneous Fission Measurements

Most measurements of spontaneous fission rates are made for uniquely characterizing new isotopes and studying the mechanisms and systematics of the decay processes of unstable nuclides. The californium isotopes, for example, were originally detected and characterized by spontaneous fission as well as by alpha decay measurements[78]. The nuclear physics literature abounds in reports of measurements and evaluations of the spontaneous fission phenomenon. The papers by Huizenga[72], and by Segre and co-workers[73,79,80] are typical.

Spontaneous fission measurement has also been used as an analytical method for determining the ^{240}Pu content of reactor produced plutonium. The application is possible because the specific spontaneous fission rate of ^{240}Pu is about 4×10^4 times greater than that of ^{239}Pu and the two isotopes make up more than 99 percent of reactor-produced plutonium. The contributions of ^{238}Pu and ^{242}Pu to the spontaneous fission count are minor and the spontaneous fission rate of ^{241}Pu is negligible. Although this method for ^{240}Pu was once used in several U.S. atomic energy project laboratories, most, if not all, isotopic analyses of plutonium are now done by surface ionization mass spectrometry. The latter method utilizes much smaller samples, is more rapid, and provides several isotopic ratios in a single measurement, rather than just the $^{240}Pu/^{239}Pu$ ratio. Nevertheless, the spontaneous fission method for ^{240}Pu provides a useful case for illustrating fission counting procedures.

C. Design of Spontaneous Fission Counters

1. Ionization Chambers

Parallel-plate ionization chambers are most commonly used for fission counting. Since the energy of a fission fragment is more than ten times that of a 5 MeV alpha particle, the necessary discrimination between the two is readily achieved by pulse height discrimination. However, nearly all nuclides that undergo spontaneous fission also decay by alpha emission with very large alpha-to-fission ratios. In the case of reactor-produced plutonium, the alpha emission rate of a source would be about 10^8 times the spontaneous fission rate. Consequently, the spontaneous fission counting system must discriminate very effectively against alpha particles to avoid the troublesome background caused by "pile-up" of alphas.

Rossi and Staub[81] treated the pile-up problem theoretically for the simplified case of a square pulse. Poisson statistics dictate the probability that (n-1) pulses will occur within the time interval, T, after a given pulse:

$$P(n-1) = \frac{(n_o T)^{n-1} e^{-n_o T}}{(n-1)!}$$

where n_o is the average number of pulses per unit time. Therefore the counting rate of pulses that have an amplitude of n times that of a single pulse will be:

$$C(n) = \frac{n_o (n_o T)^{n-1} e^{-n_o T}}{(1 + n_o T)(n-1)!}$$

The equation seriously overestimates the pile-up of alphas in fission counting, however. For example, assume a pulse length of 300 ns (a typical value for an ionization counter), a source having an alpha count rate of 10^6 c/s, and a discriminator setting such that a pile-up of six or more alpha pulses will register as a fission count. The alpha pile-up count would therefore be: $C(6) + C(7) + C(8) + \ldots$

$$= \frac{10^6}{1 + 0.3} \left(\frac{(0.3)^5}{5!} + \frac{(0.3)^6}{6!} + \frac{(0.3)^7}{7!} + \ldots \right) e^{-0.3} \approx 10 \text{ counts/s}$$

Actually, the alpha pile-up can be reduced to about 10^{-5} c/s for the above case. The discrepancy is probably largely because the pulse shape is not rectangular and linear pile-up does not occur, and the discriminator sensitivity falls off as the pile-up pulse peak becomes narrower. A theoretical treatment of other pulse shapes is quite complex, as in a recent paper by Williamson[82] showing the influence of pulse shape on the distortion of pulse height spectra by the pile-up effect.

Pile-up discrimination is achieved by using fast counters to minimize the chance pile-up of alpha pulses by narrowing the pulse width. Also, the discriminator setting is operated as high as possible and the average alpha pulse height is minimized by having a small electrode spacing in the ionization chamber. Chamber voltage and gas composition are selected to give a fast ion collection rate and RC or delay line clipping is used to shape and shorten the pulse. As a result, the counter can usually be adjusted to count nearly all fission pulses while allowing less than 10^{-4} counts/min. alpha pile-up count rate. The necessary discriminator setting is found by plotting count rate vs discriminator bias as illustrated in Figure 8. The curve of alpha pile-up counts vs discriminator setting is a straight line on semi-logarithmic graph paper. The curve is extrapolated to 10^{-4} counts / min. or less to select the desired discriminator setting. That point should be on the fission counting plateau. For any counter, a maximum acceptable

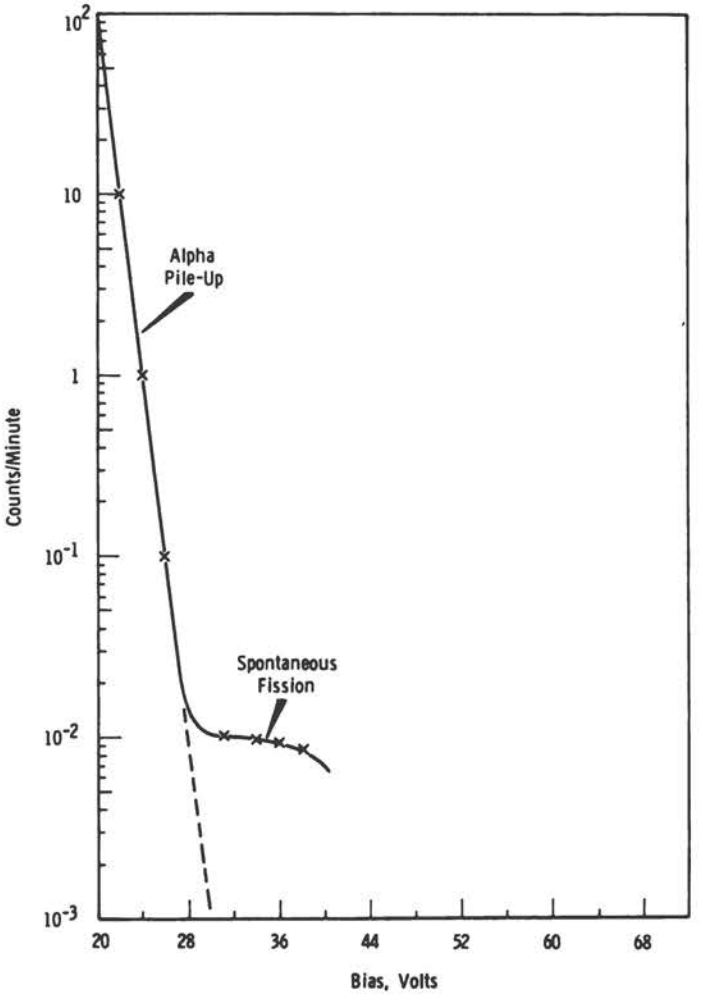

Figure 8
Typical Alpha Pile-Up Curve

alpha pile-up count rate will be found that will establish the maximum usable total alpha disintegration rate of the sample.

Ionization chambers with a screen grid shielding the collecting electrode have been used for fission counting[73,74]. A Frisch grid chamber of the type used for alpha energy analysis would serve the purpose. Gridded chambers have good alpha energy resolution since the electron pulse height is relatively insensitive to the position of the particle track. While no study appears to have been made, good energy resolution in fission counting is not likely to give any advantage because the principal problem is to differentiate between fission pulses and alpha

pile-up pulses. The differentiation is improved in the usual chamber by close electrode spacing, as explained above.

The early (pre-1945) fission counting experiments were conducted at the Los Alamos Scientific Laboratory[79,83]. Simple parallel plate chambers with spacings from 0.9 to 1.1 cm were used. Nitrogen gas filling was used initially, but pure argon was later adopted as the standard filling because the electron collection rate is higher in argon. Since the spontaneous fission count rate is normally very low, a great deal of emphasis was placed on electronic circuitry development to reduce the background or spurious count frequency and improve the discrimination against alpha counts. They were achieved by using fast rise, clipped pulses to attain good discrimination against alpha pile-up pulses, very stable, low-noise power supplies and electronic circuits, and boron-filled paraffin shields around the chambers to reduce neutron induced fissions.

White and co-workers at the Knolls Atomic Power Laboratory used spontaneous fission counters very similar to the LASL type, except that the electronics were redesigned for ac rather than battery operation[84]. Two of the KAPL fission counters were later used at Hanford[85] for ^{240}Pu measurements. In the latter work, A-CO_2 (3%) gas at about 1.5 atmospheres was used in the chambers and spontaneous fission counting of plutonium samples of up to about 2 mg was achieved with no interference from the high alpha activity. The counter backgrounds were negligible compared with the spontaneous fission count rates. The fission counter used for ^{240}Pu spontaneous fission measurements at Harwell was of a similar design[86].

Some experimental studies of counter design have been made but most of them were done on counters designed for induced fission counting, which generally gives much higher count rates and less concern for background and alpha pile-up problems. Induced fission counters are used to measure concentrations of fissile material, such as $^{235}U/^{238}U$ ratios, and for precise neutron flux measurements at low fluxes such as in reactor experiments at low power. Baer and Swift[87] tested the effects of electrode spacing and obtained good counting efficiency with electrode spacings down to about 0.4 cm with N_2 gas at atmospheric pressure. There is, however, considerable latitude available in the spacing and gas pressure used. For any particular chamber, a pressure can be found that gives an optimum fission to alpha pulse height ratio with the particular pulse shape and electronics system used. For a 1 cm electrode spacing and an argon filling, a pressure of 1.5 atmospheres was about optimum for the counters used at Hanford[85].

The gas composition in ionization counters has an effect on both the rate of electron collection and the voltage requirements and is, therefore, a design parameter that can influence pulse length. Pure nitrogen or argon can be used in spontaneous fission counters, the latter being generally preferred. However, small additions of N_2, CO_2, or CH_4 to argon increase the electron drift velocity and such mixtures have therefore been used frequently. Watt and co-workers used A-CH_4 mixtures[88], KAPL and Hanford investigators used A-CO_2(3%)[84,85], and both Nilsson and Goosey used A-N_2 mixtures[89,90]. Colli and Facchini showed the advantage of up to one percent addition of nitrogen to argon over pure argon in their study of the drift velocity of electrons in ionization chamber operation[91]. Other studies of electron drift velocities in pure argon, pure methane, and mixtures of these gases with nitrogen and carbon dioxide have been made by Bortner and co-workers[92] and by English and Hanna[93].

Absolute fission rates may be determined by coincidence counting since each fission results in two fission fragments. Back-to-back fission counter chambers are required, with the fissionable material mounted on a very thin foil between the chambers. Watt and co-workers[88] used twin hemispherical chambers with approximately 5 cm radii, argon-methane counting gas at atmospheric pressure, and circuitry that gave a pulse width of 0.5 μsec. The sources were plutonium at 2 to 20 μg/cm^2, mounted on thin plastic (40 μg/cm^2) and on thin gold (195 μg/cm^2) foils. They used the coincidence count technique to measure the spontaneous fission half-life of ^{240}Pu, obtaining the presently accepted value of $1.340 \pm 0.015 \times 10^{11}$ years.

Dual source, back-to-back, fission counters have also been used by Nilsson[89] and by Goosey[90] although not for coincidence counting, but rather for simultaneous counting of two sources of which one may be a standard. This may be advantageous in induced fission counting if the neutron source strength is not accurately reproduced from one time to another.

2. Gas Scintillation Fission Counters

Solid scintillators, both inorganic and organic, are responsive to heavy charged particles but they are not particularly well suited for fission counting because the pulse height per MeV is much smaller for a fission fragment than for an alpha particle. However, noble gases are quite suitable for scintillation counting. When the charged particle passes through the gas, it loses energy primarily by inelastic collisions, leaving a track of ions and excited atoms. The de-excitation

and recombination processes result in a burst of photons. The particularly important features of noble gas scintillation are that the decay rate of the photon pulse is very fast, of the order of millimicroseconds, and the light output is directly related to the energy of the particle absorbed and insensitive to its mass and charge. Therefore the photon pulse from a 60 MeV fission fragment of ^{240}Pu, for instance, is more than ten times greater than that from the average alpha particle. The fast pulse characteristic of the scintillation makes possible a very narrow pulse-width discrimination, therefore reducing the sensitivity to coincident or multiple pile-up pulses. A limit is imposed, however, by the fast-pulse capabilities of the electronic circuitry available.

Early experiments with gas scintillators were discussed by Eggler and Huddleston[94], and by Northrup and Nobles[95]. Application of the technique to fission counting was a natural step. Sayres and Wu in 1957[96] published a pulse spectrum of the fission fragments from ^{235}U using a xenon fission counter. For a good summary of gas scintillators, the reader is referred to Chapter 2, by R. B. Murray, in "Nuclear Instruments and Their Uses"[97].

The scintillation spectra from pure noble gases lie in the ultraviolet region. In order to measure the emission with good sensitivity, either an ultraviolet sensitive multiplier phototube must be used or a wavelength shifting material must be added. Brauer, Connally, Kinderman and Gift[98] constructed a gas scintillation fission counter and experimented with some of the wavelength shifters used by previous investigators to find a scintillation detector that gave a high amplitude pulse and at the same time was simple and convenient to operate. Their preference was for pure argon gas in conjunction with a uv-sensitive multiplier phototube (RCA type 6903 in this case). Part of the reason for that choice was that the pulse amplitude was found to be quite sensitive to variations in gas composition and maintaining a constant and reproducible gas composition is troublesome when a gas mixture is being used. In addition, the fission peak amplitude from pure argon (and the 6903 PM) was at least as good as the best from argon or helium with nitrogen or tetraphenylbutadiene as wavelength shifters.

Some of the high molecular weight organic compounds that are commonly used as wavelength shifters in liquid scintillators are also applicable to gas scintillators: diphenylstilbene, p-quaterphenyl, and 1,1,4,4-tetraphenylbutadiene are effective wavelength shifters in argon, krypton, and xenon scintillation counting[99]. Those materials may be deposited on the walls of the chamber or simply on the face of the

multiplier phototube. An optimum thickness appears to be about 20 to 30 $\mu g/cm^2$. The addition of a small amount of nitrogen to helium or argon also shifts the wavelength peak. Brauer and co-workers[98] checked the He-N_2(0.2%) system using an emission spectrograph and found the brightest emission lines from alpha scintillations to be the nitrogen lines at 3914.4 and 4269.7 angstroms.

The gas scintillation spontaneous fission counter built by Brauer and co-workers[98] had a series diode prediscriminator followed by a cathode follower preamplifier, linear amplifier, discriminator, and scaler. The system operated with about a 120 nanosecond pulse at the prediscriminator. Spontaneous fission counting of 5 mg plutonium sources was achieved with as little alpha interference as with about 2 mg sources in the ionization chamber system previously used.

D. Source Preparation

Sources for fission counting should ordinarily be prepared by vacuum evaporation or electrodeposition since other methods are not as well suited for depositing large amounts of an element in a uniformly thin and adherent form. The exceptions are the high specific fission rate nuclides, such as some of the curium and californium isotopes. Their spontaneous fission count rates are high enough that only submicrogram amounts are mounted and the deposition method is less critical. Isotopes that are very scarce are preferably mounted by electroposition in order to obtain high yields and recovery of the undeposited material[100].

For some isotopes, sources of several milligrams are needed to obtain satisfactory spontaneous fission count rates, and experimentally determined absorption corrections are made. If the sources are not uniform in thickness, self-absorption errors will be increased. In order to minimize self-absorption when mounting milligrams of an element on a fission counting source, the foils used are usually much larger than used for alpha counting, thereby spreading out the material on the foil.

Source thicknesses must be considerably less than 0.1 mg/cm^2 unless corrections for absorption loss are to be made (the range of fission fragments in metal oxides is about 10 mg/cm^2). Baer and Swift[87] tested fission counting sources at up to 2 mg/cm^2 of U_3O_8 and concluded that self-absorption was not appreciable up to 0.2 mg/cm^2, as indicated by the flatness of the fission counting plateau. These results are at variance, however, with the observations of other investigators. Alkire and co-workers[85] used plutonium deposits with average thicknesses up to 200 $\mu g/cm^2$ and their data indicate that self-absorption loss was measurable (a few percent) at a thickness of about 50 $\mu g/cm^2$. These

investigators avoided errors due to self-absorption in plutonium foils by measuring the ratio of the induced to spontaneous fission count, using standard sources of known plutonium isotopic composition to calibrate, as described below. The curves presented by White[101] indicate that a 0.1 mg/cm^2 U_3O_8 foil thickness had about a one percent self-absorption loss. Knobeloch[102] reports high absorption losses for uranium fission fragments from electrodeposited films of 32 μg/cm^2 average thickness. Since electrodeposit film uniformities are somewhat variable, absorption factors should be determined experimentally.

The electrodeposition methods for source preparation were described earlier in the section on alpha source preparations. The method of Miller and Brouns[48] has been used to plate up to 0.9 mg/cm^2 of plutonium with good appearance of the deposit. Ko plated up to 0.2 mg/cm^2 of thorium, up to 0.11 mg/cm^2 of uranium, and up to about 0.1 mg/cm^2 plutonium by his method[49]. Mitchell's method[52] is not satisfactory for those amounts of material because the high electrodeposition rate tends to produce spongy deposits. Brodsky and co-workers[50] report depositing uranium on treated aluminum foils to thickness of up to about 0.2 mg/cm^2 which were very uniform and adherent. Several other electrodeposition procedures are also reported to produce good deposits of actinide elements at up to a few tenths mg/cm^2[53,55].

Vacuum sublimation with a heated filament requires several successive filament loadings to reach the milligram level of element deposited because each filament loading is in the microgram range, due to physical limitations. To deposit large amounts, a crucible source may be used, as described by Parker[59] and by Salo and Garden[63]. Povelites reports that vacuum evaporation deposits can be made consistently more uniform than electrodeposits[103].

E. Weighing Fission Sources

The amount of sample material mounted on a fission foil must be determined in order to calculate the specific spontaneous fission count rate. Direct weighing or, in some cases, alpha counting or induced fission counting can be used advantageously. Since nearly all isotopes that undergo spontaneous fission are also alpha emitters, absolute alpha counting provides a convenient method for determining the amount of the element in the source, assuming that the specific activities and relative percentages of all alpha emitting nuclides in the source are known. Weighing by absolute alpha counting is done on a low-geometry, defined solid-angle counter by the methods discussed in the first section of this monograph.

The high fission cross-section of some nuclides allows use of neutron induced fission counting for "weighing" the source using a calibrated neutron source and counter arrangement. A normal sample of reactor-produced plutonium consists principally of the isotopes ^{239}Pu and ^{240}Pu. Alkire, Schmidt and Kinderman[85] "weighed" several sources of plutonium by microbalance, by low-geometry alpha counting, and by induced fission counting, and concluded from their data that the latter method was the most precise.

The method of induced fission counting requires the use of a standard source of known isotopic composition, but if only the ^{240}Pu - ^{239}Pu ratio is desired, the weight of plutonium on the sources does not need to be known. Only the ratio of the induced to spontaneous fission rates and the isotopic ratio are required. The relevant equations are as follows:

Let S = spontaneous fission counting rate of the sample* (cts/min)

I = induced fission counting rate of the sample* (cts/min)

SFR = specific spontaneous fission rate of ^{240}Pu (cts/min/g)

G = geometry factor for the fission counter

ϕ = thermal neutron flux for the neutron source used (neutrons/min/cm^2)

σ = ^{239}Pu fission cross-section for the above neutron source (cm^2/atom)

Then,

$$\text{grams } ^{240}Pu = \frac{S \times G}{SFR}$$

$$\text{grams } ^{239}Pu = \frac{I \times G \times 239}{(6.03 \times 10^{23}) \times \phi \times \sigma}$$

S and I in these equations should be corrected values.

Set up the ratio

$$\frac{\text{grams } ^{240}Pu}{\text{grams } ^{239}Pu} = \frac{S \times (6.03 \times 10^{23}) \times \phi \times \sigma}{I \times 239 \times SFR}$$

$$= \frac{S}{I} \times K$$

Where K is constant for a given counter and a constant neutron flux

$$K = \frac{\phi \times \sigma \times (6.03 \times 10^{23})}{239 \times SFR}$$

*About 99% of the spontaneous fissions are due to ^{240}Pu. Calculated corrections should be made to S for the contributions of ^{238}Pu and ^{242}Pu. More than 99% of the induced fissions are due to ^{239}Pu. Calculated corrections should be made for the contributions of ^{241}Pu and for the spontaneous fissions of ^{240}Pu.

One then solves experimentally for K by determining S and I for a standard source of plutonium having a known isotopic composition, obtained by mass spectrometry and alpha energy analysis. Unknown samples are counted under the same conditions as the standard and the $^{240}Pu/^{239}Pu$ ratio calculated from the S/I ratio.

The accuracy limits of the method are governed by the accuracy of the isotope composition of the standard, the accuracy of the corrections for ^{238}Pu, ^{241}Pu and ^{242}Pu contributions, and the reproducibility of the neutron flux and counting geometry between the counting of standard and samples. Tables XI and XII contain data that illustrate the sensitivity of the fission count rates to the isotopic composition of plutonium.

TABLE XI

Nuclear Data for Plutonium

Isotope	Spontaneous Fission Rate (fissions/g/min)	Thermal Neutron Fission Cross-Section (barns) (Ref. 104)
^{238}Pu	6.7×10^4	18.
^{239}Pu	0.6	746.
^{240}Pu	2.5×10^4	0.1
^{241}Pu	low	1025.
^{242}Pu	4.6×10^4	0.2

TABLE XII

Relative Contributions of "Typical" Plutonium Isotopes to Fission Count Rates

	Isotopes				
	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu
"Typical" isotopic composition (%)	0.01	93.0	6.5	0.47	0.02
Relative contribution to spont. fission rate (%)	0.41	0.03	100.	small	0.57
Relative contribution to induced fission rate (%)	0.0003	100.	0.001*	0.7	small

*Not including spontaneous fission rate.

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REFERENCES *

1. E. Regener, Ber. Physik. Ges, 6, 78-83 (1908).

2. E. Regener, Sitzb. Akad. Wiss. Berlin, 38, 948-965 (1909).

3. E. Rutherford and H. Geiger, Proc. Roy. Soc. A, 81, 141-161 (1908).

4. H. Geiger and A. Werner, Zeit. f. Phys., 21, 187-203 (1924).

5. S. Kinoshita, Proc. Roy. Soc., A, 83, 432-453 (1910).

6. E. K. Hyde, I. Perlman, and G. T. Seaborg, The Nuclear Properties of the Heavy Elements, Vol. I, Table 4.1, 212-237, Prentice-Hall, Inc., Englewood Cliffs, N. J. (1964).

7. C. M. Lederer, J. M. Hollander, and I. Perlman, Table of Isotopes, 6th ed., John Wiley & Sons, New York, N. Y.(1967).

8. W. J. Price, Nuclear Radiation Detection, 1-13, McGraw-Hill Book Co., Inc., New York, N. Y. (1958).

9. H. A. Bethe and J. Ashkin, "The Passage of Heavy Particles Through Matter," Part II in E. Segre (ed.) Experimental Nuclear Physics, Vol. 1, 166-251, John Wiley and Sons, New York, N. Y. (1953).

10. M. S. Livingston and H. A. Bethe, Rev. Mod. Phys., 9, 245-390 (1937).

11. A. H. Jaffey, "Radiochemical Assay by Alpha and Fission Measurements", Chapter 16, G. T. Seaborg and J. J. Katz, The Actinide Elements, McGraw-Hill, New York (1954).

12. N. R. Johnson, E. Eichler and G. D. O'Kelley, Nuclear Chemistry, Vol. II, Chapter VI, 85-174, Interscience Publishers, New York, (1963).

13. G. D. O'Kelley, Detection and Measurement of Nuclear Radiation, NAS-NS-3105, National Academy of Sciences, April, 1962.

14. J. E. Eichelberger, G. R. Grove, K. C. Jordan, and M. N. Wolfe, "Use of Calorimetry in Nuclear Material Accountability", in AEC and Contractor Nuclear Materials Management Symposium, October 3-5, 1962, USAEC Report TID-7660 (July, 1963).

*Reports issued by the USAEC and other agencies are available for loan at USAEC Depository Libraries, or for sale by the issuing agency. For details on availability, see The Nuclear Science Abstracts.

15. S. R. Gunn, Nucl. Instr. and Methods, 29, 1-24 (1964).

16. E. H. Fleming, Jr., A. Ghiorso and B. B. Cunningham, Phys. Rev., 88, 642-652 (1952).

17. M. L. Curtis, R. G. Olt, J. F. Eichelberger, Absolute Alpha Counting II. Standard Low Geometry Chamber, USAEC Report MLM-843 (May, 1953).

18. M. L. Curtis, J. W. Heyd, R. G. Olt, and J. F. Eichelberger, Nucleonics, 13 (No. 5), 38-44 (1955).

19. A. H. Jaffey, Rev. Sci. Inst., 25, 349-354 (1954).

20. R. F. Overman, "New Evaluation of Low Geometry Alpha Counting", USAEC Report DP-780 (Sept. 1962).

21. H. P. Robinson, "Alpha Standards", in Measurements and Standards of Radioactivity, Nuclear Science Series Report No. 24 (National Academy of Sciences Publ. No. 573) (1958).

22. A. Spernol and O. Lerch, Nucl. Instr. and Methods, 32, 293-299 (1965).

23. A. J. Cruikshank, D. J. Littler, and A. G. Ward, A Determination of η (Thermal) of Uranium-233, AECL Report CRP-378 (April 1948).

24. R. Hurst and G. R. Hall, Analyst, 77, 790-800 (1952).

25. H. P. Robinson, "A New Design for A High Precision, High Geometry α Counter", 147-154, in Metrology of Radionuclides, Proceedings of a Symposium, Organized by the IAEA, Vienna, 14-16 October 1959, International Atomic Energy Agency, Vienna 1, Austria, 1960.

26. R. A. Schneider and K. M. Harmon, eds., Analytical Technical Manual, USAEC Report HW-53368 (Feb., 1961).

27. C. M. Lederer, J. K. Poggenburg, F. Asaro, J. O. Rasmussen, and I. Perlman, Nuclear Physics, 84, 481-504 (1966).

28. W. S. Lyon and S. A. Reynolds, Nucleonics, 14 (No. 12), 44-8 (Dec., 1956).

29. E. Sakai, H. Tamura, and Y. Sakurai, J. Nucl. Sci. Tech. (Tokyo) 1, No. 3, 101-109 (1964).

30. A. Rytz, Rapport sur la Comparaison Internationale de ^{241}Am, NP-14872 (1963).

31. C. G. Bell and F. N. Hayes, (eds.) Liquid Scintillation Counting; Proceedings. Pergamon Press, New York, N. Y., 1958.

32. J. B. Birks, The Theory and Practice of Scintillation Counting, Pergamon Press, New York, N. Y., 1964.

33. J. K. Basson and J. Steyn, "Absolute Alpha Standardization with Liquid Scintillators", (letter), Proc. Phys. Soc., [London], A, 67, 297-8 (1954).

34. J. K. Basson, Anal. Chem., 28, 1472-4 (1956).

35. D. L. Horrocks and M. H. Studier, Anal. Chem., 30, 1747-50 (1958).

36. H. H. Seliger, Intern. J. Appl. Radiation and Isotopes, 8, 29-34 (1960).

37. H. Ihle, M. Karayannis, and A. Murrenhoff, "Comparative Investigations on Counting Methods for the Absolute Determination of the Activity of Alpha Emitters", JUL-298 PC, Institut fuer Physikalische Chemie, Kernforschungsanlage, Juelich, West Germany, Sept. 1965.

38. F. P. Brauer, R. W. Stromatt, J. D. Ludwick, F. P. Roberts, and W. L. Lyon, The Specific Activity and Half-Life of ^{237}Np, USAEC Report HW-59642, March 1959.

39. F. P. Brauer, R. W. Stromatt, J. D. Ludwick, F. P. Roberts, and W. L. Lyon, J. Inorg. Nucl. Chem., 12, 234-5 (1960).

40. Unpublished Data of the Chemistry and Metallurgy Division, Atomic Energy of Canada, Ltd. (1964).

41. K. M. Glover and G. R. Hall, Nature, 173, 991-2 (1954).

42. D. E. Watt, R.L.G. Keith and F. Brown, An Intercomparison of the Determination of ^{239}Pu, ^{238}Pu, ^{236}Pu by Alpha Counting Techniques in the UK, USA, and Canada, UKAEA Report AWRE-O-59/63 (July 1963).

43. M. L. Smith (ed.), Proceedings of the Seminar on the Preparation and Standardisation of Isotopic Targets and Foils, held at AERE, Harwell, on October 20 and 21, 1965, UKAEA Report AERE-R-5097 (Dec. 1965)

44. R. W. Dodson, A.C. Graves, L. Helmholz, D. L. Hufford, R. M. Potter, and J. G. Povelites, "Preparation of Foils", Chapter 1, 1-46, in Miscellaneous Physical and Chemical Techniques of the Los Alamos Project, by A. C. Graves and D. K. Fromen (eds.) National Nuclear Energy Series V-3, McGraw-Hill Book Co., New York, N. Y. (1952).

45. D. L. Hufford and B. F. Scott, "Techniques for the Preparation of Thin Films of Radioactive Material", Paper 16.1, 1149-84, in The Transuranium Elements, by G. T. Seaborg, J. J. Katz, and W. M. Manning (eds.), National Nuclear Energy Series IV-14B, McGraw-Hill Book Co., New York, N. Y. (1949).

46. K. M. Glover and P. Borrell, J. Nuclear Energy, 1, 214-17 (1955).

47. D. G. Tuck, Anal. Chim. Acta, 17, 271-4 (1957).

48. H. W. Miller and R. J. Brouns, Anal. Chem., 24, 536-8 (1952).

49. R. Ko, Nucleonics, 15, No. 1, 72-6 (1957).

50. A. Brodsky, L. W. Fagg, and T. D. Hanscome, Health Physics, 1, 189-91 (1958).

51. A. G. Samartseva, Izotopy i Izluchewiya v Khim, 318-25 (1957), available in "Transactions of the All-Union Scientific Technical Conference on the Use of Radioactive and Stable Isotopes and Radiations in the National Economy and in Science, April 4-12, 1957, Isotopes and Radiation in Chemistry", USAEC Report AE-tr-4497, 400-10. USAEC Div. of Technical Information (1961).

52. R. F. Mitchell, Anal. Chem., 32, 326-8 (1960).

53. J. Fuger, Bull. Soc. Chim. Belges., 71, 134-41 (1962).

54. D. Tomlin and G. A. Barnett, Health Physics, 11, No. 8, 783 (1965).

55. G. I. Khlebnikov and E. P. Dergunov, Atomnaya Energiya, 4, 376-7 (1958). [Soviet J. At. Energy, 4, 494-6 (1958), in English trans.].

56. M. Y. Donnan and E. K. Dukes, Anal. Chem., 36, 392-4 (1964).

57. N. Getoff and H. Bildstein, Nucl. Inst. and Methods, 36, 173-5 (1965).

58. B. D. Pate and L. Yaffe, Can. J. Chem., 34, 265-74 (1956).

59. W. Parker, Nuclear Instr. and Methods, 5, 142-7 (1959).

60. F. Asaro, F. L. Reynolds, and I. Perlman, Phys. Rev., 87, 277-85 (1952).

61. J. Milsted, The Preparation of Thin Alpha Sources by Vacuum Sublimation, UKAEA Report AERE-C/R-1379 (Feb. 1954).

62. N. Jackson, "A Vacuum Sublimation Apparatus for Preparation of Thin Sources of α-Active Materials," UKAEA Report AERE-C/R-2767 (1958); also, J. Sci. Instr., 37, 169-71 (1960).

63. A. E. Salo and N. B. Garden, The Fabrication of Radioactive Sources, USAEC Report UCRL-8928 (Oct. 1959).

64. Leon Wish, Anal. Chem., 31, 326-30 (1959).

65. N. Jackson and J. F. Short, Separation of Neptunium and Plutonium by Ion Exchange, UKAEA Report AERE-M-444 (1959).

66. S. G. Thompson and M. L. Muga, "Methods of Production and Research on Transcurium Elements", Proc. of Second UN Int. Conf. on the Peaceful Uses of Atomic Energy, 28, 331-48 (1958).

67. D. J. Carswell and J. Milsted, J. Nucl. Energy, 4, 51-4 (1957).

68. L. N. Blumberg, P. Stein and J. C. Gursky, Electrospraying of Thin Targets, USAEC Report LA-2711 (Aug. 1962).

69. M. A. Guebely and T. Sikkeland, Acta. Chem. Scand., 11, 473-8 (1957).

70. N. Bohr and J. A. Wheeler, Phys. Rev., 56, 426-50 (1939).

71. G. N. Flerov and K. A. Petrzhak, J. Phys. USSR, 3, 275-80 (1940).

72. J. R. Huizenga, Phys. Rev., 94, 158-60 (1954).

73. E. Segre and C. Weigand, Phys. Rev., 94, 157-8 (1954).

74. W. J. Whitehouse and W. Galbraith, Phil. Mag., 31, 429-43 (1950).

75. G. Goldstein and S. A. Reynolds, Nuclear Data, A, (No. 5), 1, 435-52 (1966).

76. Reactor Physics Constants, Table 1-25, ANL-5800, 2nd Ed., USAEC Div. of Tech. Information (July 1963).

77. A. B. Smith, P. R. Fields and A. M. Friedman, Phys. Rev., 106, 779-81 (1957).

78. H. Diamond, L. B. Magnusson, J. F. Mech, C. M. Stevens, A. M. Friedman, M. H. Studier, P. R. Fields, and J. R. Huizenga, Phys. Rev., 94, 1083-4 (1954).

79. E. Segre, Phys. Rev., 86, 21-8 (1952).

80. O. Chamberlain, G. W. Farwell, and E. Segre, Phys. Rev., 94, 156 (1954).

81. B. B. Rossi and H. H. Staub, Ionization Chambers and Counters, 203-19, McGraw-Hill Book Co., 1949.

82. J. H. Williamson, "Distortion of Pulse Height Spectra by Pile-Up Effect", Rev. Sci. Inst., 37 (No. 6), 736-9 (1966).

83. G. W. Farwell, E. Segre, A. Spano, and C. E. Wiegand, Further Investigations of Spontaneous Fission in Plutonium, USAEC Report LA-490 (April 1946).

84. W. R. Kanne, F. A. White, I. H. Dearnley, and S. B. Dunham, unpublished data, Knolls Atomic Power Laboratory, General Electric Co. (1948).

85. G. J. Alkire, H. R. Schmidt, and E. M. Kinderman, The Fission Counter: Its Use in Plutonium Isotopic Analysis, USAEC Report HW-23486 (February 1952).

86. W. Galbraith, K. M. Glover, G. R. Hall, and W. J. Whitehouse, The Spontaneous Fission Rate of ^{240}Pu, UKAEA Report AERE-A/R-814 (April 1951).

87. W. Baer and O. F. Swift, Rev. Sci. Inst., 23, 55-6 (1952).

88. D. E. Watt, F. J. Bannister, J. B. Laidler and F. Brown, Phys. Rev., 126, 264-5 (1962).

89. G. Nilsson, Nuclear Instr. and Methods, 13, 183-9 (1961).

90. M. H. Goosey, A Fission Counter for the Routine Isotopic Analysis of Plutonium, USAEC Report DP-552 (March 1961), (Declass., Aug. 1966).

91. L. Colli and U. Facchini, Rev. Sci. Inst., 23, 39-42 (1952).

92. T. E. Bortner, G. S. Hurst and W. G. Stone, Rev. Sci. Inst., 28, 103-8 (1957).

93. W. N. English and G. C. Hanna, Can. J. Phys., 31, 768-97 (1953).

94. C. Eggler and C. M. Huddleston, Nucleonics, 14, No. 4, 34-6 (1956).

95. J. A. Northrup and R. Nobles, Nucleonics, 14, No. 4, 36-7 (1956).

96. A. Sayres and C. S. Wu, Rev. Sci. Inst., 28, 758-64 (1957).

97. A. H. Snell (ed.), Nuclear Instruments and Their Uses, Vol. I, 82-165, John Wiley and Sons, Inc., New York, N. Y. (1962).

98. F. P. Brauer, R. E. Connally, E. M. Kinderman, and M. W. Gift, unpublished data, Hanford Laboratories, General Electric Co. (1957).

99. J. A. Northrup, Rev. Sci. Inst., 29, 437-8 (1958).

100. H. L. Smith, Los Alamos Scientific Laboratory, New Mexico, private communication, December 1967.

101. P. H. White, "Alpha and Fission Counting of Thin Foils of Fissile Material", paper 29, 157-162, in Proceedings of the Seminar on the Preparation and Standardisation of Isotopic Targets and Foils, (Harwell, Oct. 20-21, 1965), AERE-R-5097 (Dec. 1965).

102. G. W. Knobeloch, Los Alamos Scientific Laboratory, private communication, December 1967.

103. J. G. Povelites, "Preparation of Thin Films of UO_2 and PuO_2 by Vacuum Evaporation", paper 5, 23-28, Proceedings of the Seminar on the Preparation and Standardisation of Isotopic Targets and Foils, (Harwell, Oct. 20-21, 1965), AERE-R-5097 (Dec. 1965).

104. D. J. Hughes and R. B. Schwartz, Neutron Cross Sections, USAEC Report BNL-325, 2nd Ed. (1958), and Supplements (1960 and 1966).

APPENDIX A

PROCEDURES FOR PREPARATION OF ALPHA AND FISSION SOURCES

Several representative electrodeposition procedures are given here for the convenience of the reader. Examination of the original papers is urged, however, for the benefit of a more complete discussion of each procedure. The procedures are generally used for deposition of very small amounts of material on foils having an area of a few square centimeters. When applied to spontaneous fission counting, microgram to milligram amounts of the nuclide are often desired, calling generally for some modification of the electrodeposition procedure, primarily as follows:

(1) Use of much larger foils to spread the source material.

(2) Use of longer plating times and possibly a series of successive platings with foil ignition and hand polishing of the deposit between steps.

(3) Increased stirring efficiency. A reciprocating stirrer using a perforated anode is more effective than a rotating anode.

Excellent sources can also be prepared by vacuum sublimation and electrospraying methods. The procedures used are to a large extent unique to the particular apparatus at hand. For descriptions of apparatus and procedures, references 43, 61, 59, 67, and 68 are recommended.

1. PREPARATION OF URANIUM SOURCES BY ELECTRODEPOSITION

Procedure A (from reference 50), for depositing up to about 200 µg of uranium.

1. Add the chosen amount of uranium ion solution to a dilute nitric or nitric and perchloric acid solution and adjust the volume to about 15 mℓ.
2. Neutralize to the blue endpoint of bromcresol green using 1 M NH_4OH solution and back titrate to the green endpoint (pH of 5-6 using 0.1 M HNO_3).
3. Add the electrolyte to an electrodeposition cell having a capacity of at least 30 mℓ. A cell similar to one of those shown previously in Figures 6 and 7 is suitable.
4. Add 5 mℓ of 0.4 M ammonium acetate solution.
5. Place the cell in a water bath at 85-90°C.

6. Electrolyze for four hours with the following conditions:
   (a) keep the water bath at 85-90°C;
   (b) stir vigorously; e.g., operate a rotating disc stirrer at about 300 rpm;
   (c) keep the cathode current density at about 100 ma/cm^2 (the cell voltage was 5-6 volts with the apparatus described in reference 50. The value will be affected by electrode areas and electrolyte concentrations).
7. After deposition, drain the cell and rinse quickly with distilled water, remove the freshly deposited source, rinse again with distilled water, and dry for 20 minutes under a heat lamp. With platinum foils, ignite the foil for five minutes at about 800°C to dehydrate the deposit. With other foil materials, lengthen the period of drying with a heat lamp.

Procedure B. (From M. R. Weiler and W. Y. Matsumoto, "Determination of ^{232}U and ^{233}U in Irradiated Thorium", USAEC Report BNWL-159, Nov. 1965).

1. Transfer the uranium, previously purified by ether extraction, to 10 ml of solution that is 1 N in NH_4NO_3 and 0.1 N in HNO_3.
2. Adjust the solution to a pH of 1.0 to 1.5 with dilute NH_4OH.
3. Transfer to an electrodeposition cell containing the desired foil as the cathode and a platinum stirrer-anode.
4. Electrolyze at 100 ma/cm^2 with continuous stirring until a pH of 8 or greater is reached.
5. Remove the cathode, rinse with distilled water, and ignite to a dull red heat in a flame.

2. ELECTRODEPOSITION OF PLUTONIUM AND OTHER ACTINIDES

Procedure A. (From reference 49).

1. Carry out the electrodeposition in a cell similar to those described previously (see Figures 6 and 7). Use platinum anode-stirrer rotating about 200 rpm at a distance of about 1 cm above the cathode.
2. Prepare the necessary supporting electrolyte in each case, add it to the cell, turn on the stirrer, and then add from

TABLE I - ACTINIDE PROCEDURE A

Element	Amount*	Supporting Electrolyte	Cathode Current Density	Duration of Electrolysis
Thorium	3 mg per 11 cm^2 of Cu of Pt	0.05 M H_2SO_4 - 0.08 M NH_4COOH	4.5 ma/cm^2	16 hr
Uranium	0.7 mg/6 cm^2 of Cu or passivated Pt	0.2 M $HClO_4$ - 0.15 M NH_4COOH	13-16 ma/cm^2	1 hr
Neptunium	2.5 $\mu g/cm^2$ on Cu or passivated Pt	0.2 M $HClO_4$ - 0.15 M NH_4COOH	50-60 ma/cm^2	1 hr
Plutonium	Up to 115 $\mu g/1.5$ cm^2 on stainless steel	0.05 M H_2SO_4 - 0.076 M NH_4COOH or 0.2 M $HClO_4$ - 0.15 M NH_4COOH	30 ma/cm^2 50-60 ma/cm^2	1 to 4 hrs
Americium and Curium	Up to 5.3 $\mu g/1.5$ cm^2 on Pt, Cu, or SS	0.2 M HCOOH - 0.15 M NH_4COOH	50-60 ma/cm^2	2 hr

*Maximum amount deposited and cathode material used in the tests reported in reference 49.

a pipet the desired amount of a solution of the nuclide to be deposited. The volume of the solution added should not exceed 10 percent of the volume of supporting electrolyte.

3. Carry out the electrolysis for each nuclide as specified in Table I.
4. At the end of the electrolysis, siphon out the electrolyte, rinse the cell with dilute NH_4OH and then acetone. Disassemble the cell, rinse again with acetone, and dry thoroughly under a heat lamp. Platinum discs may be flame-dried or heated to red heat in a furnace.

Procedure B. (From reference 51).

1. For thorium, uranium, plutonium or americium in a nitric, hydrochloric, or perchloric acid solution.
   a. Adjust the volume of the electrolyte containing the desired amount of the nuclide to be deposited to about 20 mℓ.
   b. Adjust to a pH of 2.5 to 3.0 by adding NH_4OH.
   c. Place in a suitable cell with a platinum anode-stirrer and the desired foil as the cathode. Electrolyze for two hours at a 100 ma/cm^2 cathode current density.
   d. Drain the cell, rinse with distilled water, remove the foil and rinse again, and dry thoroughly under a heat lamp. If a platinum foil is used, it may be ignited for a few minutes.
2. For neptunium in a nitric, hydrochloric, or perchloric acid solution.
   a. Adjust the volume of electrolyte to about 80 mℓ and adjust the pH to 3.0 by adding NH_4OH.
   b. Place in a suitable cell for electrodeposition on the desired foil and electrolyze for 10 minutes at a cathode current density of about 250 ma/cm^2.
   c. Complete the deposition as in step (d) above.

Procedure C. (Adapted from reference 52). Rapid Electrodeposition of Actinides.

The actinides are plated from a dilute $NH_4Cℓ$-HCℓ solution onto platinum disk cathodes with an exposed area of about 3 cm^2. The following

conditions resulted in high yields for Th, Pa, U, Np, Pu, Am and Cm in 15-minute electrodepositions:

(1) Adjust the electrolyte volume to 4 to 5 mℓ.
(2) Adjust the pH to about 1 with NH_4OH and HCℓ.
(3) Have the final $Cℓ^-$ concentration about 0.1 to 0.2 g/mℓ.
(4) Electrolyze with a current of about 2 amperes (about 0.7 amp/cm^2).
(5) Have the anode-stirrer located about 0.5 cm above the cathode and rotating rapidly.

Note: Use a lower current density for plating more than submicrogram amounts to avoid poorly adherent deposits.

APPENDIX B
TESTING OF COUNTERS AND COUNTING STATISTICS

The fundamentals of counting statistics are discussed in most radiochemistry textbooks. A few procedures given here are particularly useful in high accuracy counting and are generally not described in textbooks in a way that the average radiochemist can apply them.

1. DETERMINATION OF THE OPERATING RANGE OF AN ALPHA COUNTER

In general the operating point is on the voltage plateau between one-third and one-half of the way up the plateau. The setting must be consistent with the calibration. The plateau should be checked daily and the voltage setting posted for all users of the instrument to follow. A changing plateau position is a warning of electronic or detector malfunctioning.

The measurement of the voltage plateau is made as follows:

1. Place a high quality standard or test source with a count rate of about 50,000 c/m in the standard counting position.
2. Turn the detector voltage up to a point at which alpha counting just starts. Record the count rate at this point and at 50 volt intervals between the threshold and the point at which the count rate begins increasing markedly. (For a ZnS scintillation counter, use 25-volt intervals).
3. Plot the count rate vs voltage and choose the operating point.

2. DETERMINATION OF THE COINCIDENCE LOSS CORRECTION OF A COUNTER BY THE PAIRED-DISC METHOD

The coincidence-loss correction of a counter is made using the formula:

$$N = R + R^2T$$

where

N = true counting rate

R = observed counting rate

T = the dead time of the counter

The formula is sufficiently accurate without the higher terms in T that have been dropped. The simplest procedure for measuring T is the paired disc method described here*. Estimates of T made by a double-pulse generator or an oscilloscope method are less accurate. Another accurate method of measuring counter dead time is reported by J. P. Balagna**. It involves following the decay of a very active short-lived alpha source such as 60-minute ^{212}Bi-^{212}Po (or 54-minute ^{116}In in the case of β counters) from very high count rates where coincidence losses are significant to low count rates where losses are negligible. A computer program is used to solve for the counter dead time required to linearize the single component exponential decay time.

The Paired Disc Procedure

1. Cut a sample mounting foil in halves and carefully flatten the two halves.
2. Mount sufficient alpha emitter on each half foil to give an alpha activity on each foil of 30,000 to 40,000 counts per minute. The amount of activity on the foils does not need to be known exactly. The deposits should be thin, uniform, and relatively solids-free so that the counting plateaus are quite flat.
3. Count the foils in the standard counting position in the following sequence:
   a. count disc A for 10 minutes
   b. count discs A + B for 10 minutes
   c. count disc B for 10 minutes
   d. repeat the sequence

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*T. P. Kohman, Anal. Chem., 21, 352-64 (1949).

**J. P. Balagna, Los Alamos Scientific Laboratory, Los Alamos, New Mexico, private communication, December 1967.

4. Calculate the counter dead time using the formula:

$$T = \frac{R_A + R_B - R_{AB} - b}{\sqrt{R^2_{AB} - R^2_A - R^2_B}}$$

where:

R_A = average count rate of disc A

R_B = average count rate of disc B

R_{AB} = average count rate of disc A + disc B

b = background count rate

5. Coincidence corrections are made by applying the simple formula:

$$N = R + TR^2$$

where

N = the true count rate

R = the observed count rate

T = the dead time of the counter

Comments:

The specified count rates and counting times are appropriate for counters with values of T of about 0.1 μmin. If appreciably larger or smaller values of T are found, the source strengths of the half-discs may be adjusted accordingly. The relative standard error of the estimate of T will be 10 to 15 percent for $T = 10^{-7}$ min., which is sufficiently precise for most cases. The relative standard error of the T measurement may be calculated using the following approximation:

$$\frac{\sigma T}{T} = \left(\frac{R_A/t_a + R_B/t_b + R_{AB}/t_{ab}}{(R_A + R_B - R_{AB})^2} \right)^{1/2}$$

3. MAKING THE CHI-SQUARE TEST FOR COUNTER STABILITY

The Chi-Square test is used to show that the variations between successive counts of a given source are no greater than expected from counting statistics.

Procedure:

1. Place a test source having an activity of about 50,000 c/m in the standard counting position.
2. Count the disc five times for 10-minute intervals. Record

the values, calculate the counting rates and the mean counting rate, and calculate the χ^2 as follows:

3. Calculation:

$$\chi^2 = \frac{\sum_{i=1}^{n}(C_i - \bar{C})^2}{\bar{C}}$$

n = number of trials (=5)

C_i = total counts observed in the $i^{\underline{th}}$ trial

$\bar{C}$ = mean value $\left(= \sum_{i=1}^{5} \frac{C_i}{5} \right)$

One may prefer to use the form:

$$\chi^2 = \frac{\sum_{i=1}^{n} C_i^2 - n\bar{C}^2}{\bar{C}}$$

4. Compare the χ^2 with the tabulated values for a 99% probability at 4 degrees of freedom. If the χ^2 is outside the 99% limit, re-run the test before considering the instrument out of control. (For more stringent control, one may prefer to use the 95% limit).

The limits for the χ^2 with 4 degrees of freedom are:

at 99% probability: 0.297 and 13.3

at 95% probability: 0.711 and 9.49

Notes: The calculation may be made using count rates rather than the total counts. In this case the counting time, t, appears in the formula:

$$\chi^2 = \frac{\sum_{i=1}^{n} (x_i - \bar{x})^2}{\bar{x}/t}$$

Example: Five 10-minute counts were taken as follows and tabulated along with the calculations:

n	C_i	$(C_i - \bar{C})$	$(C_i - \bar{C})^2$
1	524,610	-248	61,504
2	525,050	+192	36,864
3	524,550	-308	94,864
4	524,210	-648	419,904
5	525,870	+1012	1,024,144
ΣC_i =	2,624,290		SS = 1,637,280

$\frac{\Sigma C_i}{5} = \bar{C} = 524,858$

$\chi^2 = \frac{SS}{\bar{C}} = 3.12$

Since this value of χ^2 is within the control limits, the counter is in good control; i.e., the variations observed between counts can be attributed entirely to the counting statistics.

4. CALCULATING THE CONFIDENCE LIMITS OF A COUNT

The 95% and 99% confidence limits for total counts of less than about 50 are shown graphically in Figure 1. When the total counts observed exceed the range of the graph the confidence limits can be caluclated as follows:

$$\sqrt{\mu_2} = \sqrt{C + 1} + \frac{f}{2}$$

$$\sqrt{\mu_1} = \sqrt{C} - \frac{f}{2}$$

where

μ_2 = the upper confidence limit

μ_1 = the lower confidence limit

C = the total counts observed
(= Σx_i or $t\bar{x}$)

f = 1.96 for the 95% confidence limits

= 2.58 for the 99% confidence limits

A simple approximation of the limits is obtained by using the formula:

$$\mu = C \pm f\sqrt{C}$$

To obtain the confidence limits of a count rate or a mean value, simply divide each limit by the count time. Thus, where $\bar{x} = \frac{C}{t}$, the upper and lower limits are μ_2/t and μ_1/t, respectively.

5. A TEST OF WHETHER TWO SOURCES HAVE THE SAME MEAN COUNT RATE

The difference between the observed count rates should fall within the probability limits given by the "normal" distribution if the source strengths are not significantly different. A test of this may be made as follows:

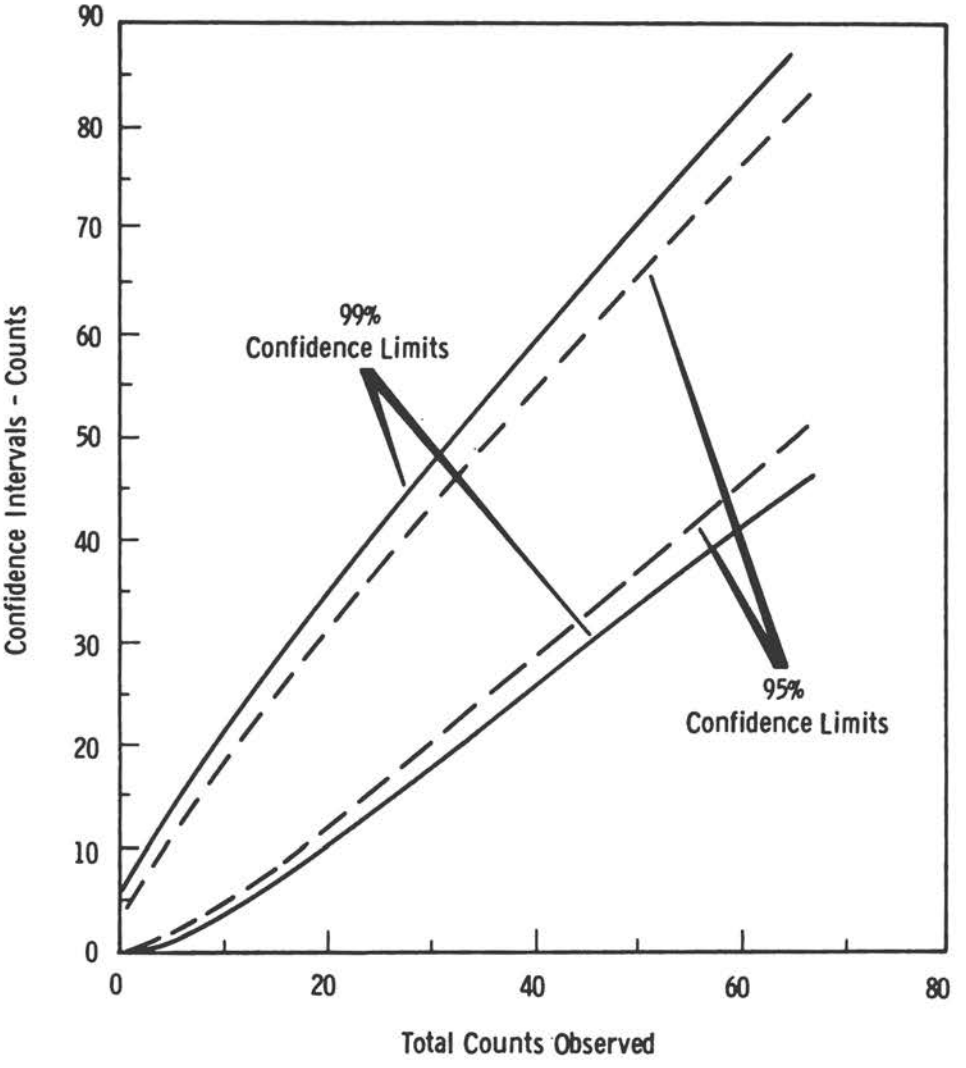

Figure 1.
(Appendix B)
Poisson Distribution Confidence Limits

1. Suppose mean count rates of $\bar{x}_1$ and $\bar{x}_2$ are obtained for the two sources in total counting times t_1 and t_2, respectively.

2. Solve for the overall mean:

$$\hat{\mu} = \frac{t_1\bar{x}_1 + t_2\bar{x}_2}{t_1 + t_2}$$

3. Calculate two parameters, p_1 and p_2, as follows:

$$p_1 = 2\left(\sqrt{t_1\bar{x}_1 + 1} - \sqrt{t_1\hat{\mu}}\right)$$

$$p_2 = 2\left(\sqrt{t_2\bar{x}_2} - \sqrt{t_2\hat{\mu}}\right)$$

4. If the sources are not significantly different, the differences between p_1 and p_2 should be less than the following range limits:

   For a 5% level of significance: 2.77

   For a 1% level of significance: 3.64

   Note: This test may also be used to compare two counters.

   Example: Suppose a given standard alpha source was counted on two instruments with the following results. Do the results indicate that the two counters have different geometry factors?

Counter	Count Rate	Total Counting Time
1	10,000 c/m	45 min.
2	10,050 c/m	30 min.

$$\hat{\mu} = \frac{450{,}000 + 301{,}500}{45 + 30} = 10{,}020 \text{ c/m}$$

$$p_1 = 2\left(\sqrt{450001} - \sqrt{450900}\right) = -1.44$$

$$p_2 = 2\left(\sqrt{301500} - \sqrt{300600}\right) = 1.64$$

$$p_2 - p_1 = 3.08$$

Therefore, the data indicate that the counters have different geometry factors at the 95% confidence level. Additional counting should be done to confirm this and obtain an improved estimate of the normalization factor between the counters.