



Designation: D3648 – 04

# Standard Practices for the Measurement of Radioactivity<sup>1</sup>

This standard is issued under the fixed designation D3648; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon ( $\epsilon$ ) indicates an editorial change since the last revision or reapproval.

## 1. Scope

1.1 These practices cover a review of the accepted counting practices currently used in radiochemical analyses. The practices are divided into four sections:

	Section
General Information	6 to 11
Alpha Counting	12 to 22
Beta Counting	23 to 33
Gamma Counting	34 to 41

1.2 The general information sections contain information applicable to all types of radioactive measurements, while each of the other sections is specific for a particular type of radiation.

1.3 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

## 2. Referenced Documents

### 2.1 ASTM Standards:<sup>2</sup>

D1066 Practice for Sampling Steam

D1129 Terminology Relating to Water

D1943 Test Method for Alpha Particle Radioactivity of Water

D2459 Test Method for Gamma Spectrometry of Water<sup>3</sup>

D3084 Practice for Alpha-Particle Spectrometry of Water

D3085 Practice for Measurement of Low-Level Activity in Water<sup>3</sup>

D3370 Practices for Sampling Water from Closed Conduits

D3649 Practice for High-Resolution Gamma-Ray Spectrometry of Water

E380 Practice for Use of the International System of Units (SI) (the Modernized Metric System)

<sup>1</sup> These practices are under the jurisdiction of ASTM Committee D19 on Water and are the direct responsibility of D19.04 on Methods of Radiochemical Analysis.

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<sup>2</sup> For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

<sup>3</sup> Withdrawn.

### 2.2 ANSI/ISO Standards:<sup>4</sup>

ANSI N42.14 Calibration and Use of Germanium Spectrometers for the Measurement of Gamma-Ray Emission Rates of Radionuclides

ISO Guide to the Expression of Uncertainty in Measurement, 1993

## 3. Terminology

### 3.1 Definitions:

3.1.1 For definitions of terms used in these practices, refer to Terminology D1129. For an explanation of the metric system, including units, symbols, and conversion factors, see Practice E380.

## 4. Summary of Practices

4.1 The practices are a compilation of the various counting techniques employed in the measurement of radioactivity. The important variables that affect the accuracy or precision of counting data are presented. Because a wide variety of instruments and techniques are available for radiochemical laboratories, the types of instruments and techniques to be selected will be determined by the information desired. In a simple tracer application using a single radioactive isotope having favorable properties of high purity, energy, and ample activity, a simple detector will probably be sufficient and techniques may offer no problems other than those related to reproducibility. The other extreme would be a laboratory requiring quantitative identification of a variety of radionuclides, preparation of standards, or studies of the characteristic radiation from radionuclides. For the latter, a variety of specialized instruments are required. Most radiochemical laboratories require a level of information between these two extremes.

4.2 A basic requirement for accurate measurements is the use of accurate standards for instrument calibration. With the present availability of good standards, only the highly diverse radiochemistry laboratories require instrumentation suitable for producing their own radioactive standards. However, it is advisable to compare each new standard received against the previous standard.

<sup>4</sup> Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036.

4.3 Thus, the typical laboratory may be equipped with proportional or Geiger-Mueller counters for beta counting, sodium iodide or germanium detectors, or both, in conjunction with multichannel analyzers for gamma spectrometry, and scintillation counters suitable for alpha- or beta-emitting radionuclides.

## 5. Significance and Use

5.1 This practice was developed for the purpose of summarizing the various generic radiometric techniques, equipment, and practices that are used for the measurement of radioactivity.

## GENERAL INFORMATION

### 6. Experimental Design

6.1 In order to properly design valid experimental procedures, careful consideration must be given to the following:

- 6.1.1 radionuclide to be determined,
- 6.1.2 relative activity levels of interferences,
- 6.1.3 type and energy of the radiation,
- 6.1.4 original sample matrix, and
- 6.1.5 required accuracy.

6.2 Having considered 6.1.1-6.1.5, it is now possible to make the following decisions:

- 6.2.1 chemical or physical form that the sample must be in for radioassay,
- 6.2.2 chemical purification steps,
- 6.2.3 type of detector required,
- 6.2.4 energy spectrometry, if required,
- 6.2.5 length of time the sample must be counted in order to obtain statistically valid data,
- 6.2.6 isotopic composition, if it must be determined, and
- 6.2.7 size of sample required.

6.3 For example, gamma-ray measurements can usually be performed with little or no sample preparation, whereas both alpha and beta counting will almost always require chemical processing. If low levels of radiation are to be determined, very large samples and complex counting equipment may be necessary.

6.3.1 More detailed discussions of the problems and interferences are included in the sections for each particular type of radiation to be measured.

### 7. Apparatus

#### 7.1 Location Requirements:

7.1.1 The apparatus required for the measurement of radioactivity consists, in general, of the detector and associated electronic equipment. The latter usually includes a stable power supply, preamplifiers, a device to store or display the electrical pulses generated by the detector, or both, and one or more devices to record information.

7.1.2 Some detectors and high-gain amplifiers are temperature sensitive; therefore, changes in pulse amplitude can occur as room temperature varies. For this reason, it is necessary to provide temperature-controlled air conditioning in the counting room.

7.1.3 Instrumentation should never be located in a chemical laboratory where corrosive vapors will cause rapid deterioration and failure.

7.2 *Instrument Electrical Power Supply*—Detector and electronic responses are a function of the applied voltage; therefore, it is essential that only a very stable, low-noise electrical supply be used or that suitable stabilization be included in the system.

#### 7.3 Shielding:

7.3.1 The purpose of shielding is to reduce the background count rate of a measurement system. Shielding reduces background by absorbing some of the components of cosmic radiation and some of the radiations emitted from material in the surroundings. Ideally, the material used for shielding should itself be free of any radioactive material that might contribute to the background. In practice, this is difficult to achieve as most construction materials contain at least some naturally radioactive isotopes (such as  $^{40}\text{K}$ , members of the uranium and thorium series, and so forth). The thickness of the shielding material should be such that it will absorb most of the soft components of cosmic radiation. This will reduce cosmic-ray background by approximately 25 %. Shielding of beta- or gamma-ray detectors with anticoincidence systems can further reduce the cosmic-ray or Compton scattering background for very low-level counting.

7.3.2 Detectors have a certain background counting rate from naturally occurring radionuclides and cosmic radiation from the surroundings; and from the radioactivity in the detector itself. The background counting rate will depend on the amounts of these types of radiation and on the sensitivity of the detector to the radiations.

7.3.3 In alpha counting, low backgrounds are readily achieved since the short range of alpha particles in most materials makes effective shielding easy. Furthermore, alpha detectors are quite insensitive to the electromagnetic components of cosmic and other environmental radiation.

#### 7.4 Care of Instruments:

7.4.1 The requirements for and advantages of operating all counting equipment under conditions as constant and reproducible as possible have been pointed out earlier in this section. The same philosophy suggests the desirability of leaving all counting equipment constantly powered. This implies leaving the line voltage on the electrical components at all times. The advantage to be gained by this practice is the elimination of the start-up surge voltage, which causes rapid aging, and the instability that occurs during the time the instrument is coming up to normal temperature.

7.4.2 A regularly scheduled and implemented program of maintenance is helpful in obtaining satisfactory results. The maintenance program should include not only checking the necessary operating conditions and characteristics of the components, but also regular cleaning of the equipment.

7.5 *Sample and Detector Holders*—In order to quantify counting data, it is necessary that all samples be presented to the detector in the same “geometry.” This means that the samples and standards should be prepared for counting in the same way so that the distance between the source and the detector remains as constant as possible. In practice, this

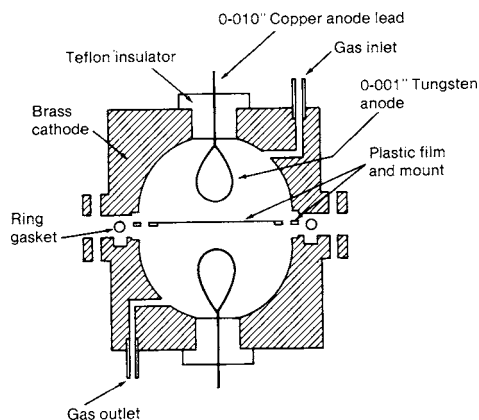


FIG. 1 The 4- $\pi$ -Counting Chamber

usually means that the detector and the sample are in a fixed position. Another configuration often used is to have the detector in a fixed position within the shield, and beneath it a shelf-like arrangement for the reproducible positioning of the sample at several distances from the detector.

**7.6 Special Instrumentation**—This section covers some radiation detection instruments and auxiliary equipment that may be required for special application in the measurement of radioactivity in water.

#### 7.6.1 4- $\pi$ Counter:

**7.6.1.1** The 4- $\pi$  counter is a detector designed for the measurement of the absolute disintegration rate of a radioactive source by counting the source under conditions that approach a geometry of 4- $\pi$  steradians. Its most prevalent use is for the absolute measurement of beta emitters (1, 2).<sup>5</sup> For this purpose, a gas-flow proportional counter similar to that in Fig. 1 is common. It consists of two hemispherical or cylindrical chambers whose walls form the cathode, and a looped wire anode in each chamber. The source is mounted on a thin supporting film between the two halves, and the counts recorded in each half are summed. A 10 % methane-90 % argon gas mixture can be used; however, pure methane gives flatter and longer plateaus and is preferred for the most accurate work. The disadvantage is that considerably higher voltages, about 3000 V, rather than the 2000 V suitable for methane-argon, are necessary. As with all gas-filled proportional counters, very pure gas is necessary for very high detector efficiency. The absence of electronegative gases that attach electrons is particularly important since the negative pulse due to electrons is counted in this detector. Commercial chemically pure (cp) gases are ordinarily satisfactory, but they should be dried for best results. A high-voltage power supply for the detector, an amplifier, discriminator, and a scaler complete the system.

**7.6.1.2** To convert counting rate to disintegration rate, the principal corrections required are for self-absorption in the source and for absorption in the support film. The support film should be as thin as practicable to minimize absorption of beta particles emitted in the downward direction. Polyester film

with a thickness of about 0.9 mg/cm<sup>2</sup> is readily available and easily handled. However, it is too thick for accurate work with the lower energy beta emitters. For this purpose, thin films ( $\approx$ 5 to 10  $\mu$ g/cm<sup>2</sup>) are prepared by spreading a solution of a polymer in an organic solvent on water. VYNS (1), Formvar (2), and Tygon (3) plastics have been used for this purpose.

**7.6.1.3** The films must be made electrically conducting (since they are a part of the chamber cathode) by covering them with a thin layer (2 to 5  $\mu$ g/cm<sup>2</sup>) of gold or palladium by vacuum evaporation. The absorption loss of beta particles in the film must be known. Published values can be used, if necessary, but for accurate work an absorption curve using very thin absorbers should be taken (1). The “sandwich” method, in which the film absorption is calculated from the decrease in counting rate that occurs when the source surface is covered with a film of the same thickness as the backing film, is suitable for the higher beta energies.

**7.6.1.4** The source itself must be very thin and deposited uniformly on the support to obtain negligible self-absorption. Various techniques have been used for spreading the source; for example, the evaporation of <sup>63</sup>Ni-dimethylglyoxime onto the support film (1), the addition of a TFE-fluorocarbon suspension (3), colloidal silica, or insulin to the film as spreading agents, and hydrolysis (2). Self-absorption in the source or mount can be measured by 4- $\pi$  beta-gamma coincidence counting (4, 5). The 4- $\pi$  beta counter is placed next to a NaI(Tl) detector, or a portion of the chamber wall is replaced by a NaI(Tl) detector, and the absolute disintegration rate is evaluated by coincidence counting (6, 7). By adding a suitable beta-gamma tracer, the method has been used for pure beta as well as beta-gamma emitters (8). Accurate standardization of pure low-energy beta emitters (for example, <sup>63</sup>Ni) is difficult, and the original literature should be consulted by those inexperienced with this technique.

**7.6.1.5** Photon (gamma and strong X-ray) scintillation counters with geometries approaching 4- $\pi$  steradians can be constructed from NaI(Tl) crystals in either of two ways. A well crystal (that is, a cylindrical crystal with a small axial hole covered with a second crystal) will provide nearly 4- $\pi$  geometry for small sources, as will two solid crystals placed very close together with a small source between them. The counts from both crystals are summed as in the gas-flow counter. The deviation for 4- $\pi$  geometry can be calculated from the physical dimensions. For absolute gamma-ray counting, the efficiency of the crystal for the gamma energy being measured and the absorption in the crystal cover must be taken into account. Additional information on scintillation counting is given in 7.6.4. The liquid scintillation counter is also essentially a 4- $\pi$  counter for beta particles, since nearly all the radiations are emitted into and interact with the detecting medium.

**7.6.2 Low-Geometry Counters**—This type of instrument is particularly useful for the absolute counting of alpha particles. The alpha emitter, in the form of a very thin solid source, is placed at a distance from the detector such that only a small fraction (<1 %) of the alpha particles are emitted in a direction to enter the counter. This solid angle is obtained from the physical measurements of the instrument. The space between the source and the detector is evacuated to eliminate the loss of

<sup>5</sup> The boldface numbers in parentheses refer to the list of references appended to these practices.



alpha particles by absorption in air. The detector can be any counter that is 100 % efficient for all alpha particles that enter the sensitive volume—a gas-flow proportional counter with a window that is thin (less than approximately 1 mg/cm<sup>2</sup>) compared to the range of the alpha particles or the semiconductor alpha detector with a 1-mg/cm<sup>2</sup> covering. The advantages of this instrument for absolute alpha counting are that: (1) the effect of absorption of alpha particles in the source itself is kept to a minimum since only particles that travel the minimum distance in the source enter the detector (particles that have longer paths in the source are emitted at the wrong angle, and (2) backscattered alpha particles (those that are emitted into the source backing and are reflected back up through the source) lose sufficient energy so that they cannot enter the detector. One such instrument is described in Reference (9).

### 7.6.3 Internal Gas Counters:

7.6.3.1 The internal gas counter is so named because the radioactive material, in the gaseous state, is placed inside a counting chamber and thus becomes part of the counting gas itself. It is useful for high-efficiency counting of weak beta- and X-ray emitters. The radiations do not have to penetrate a counter window or solid source before entering the sensitive volume of a detector. The counter may be an ionization chamber, or it may be operated in the Geiger-Mueller or proportional mode. Most present-day instruments are of the latter type, and they generally take the form of a metal or metal-coated glass cylinder as a cathode with a thin anode wire running coaxially through it and insulated from the cylinder ends. A wire through the wall makes electrical contact to the cathode. The counter has a tube opening through which it may be connected to a gas-handling system for filling. The purity of the gas is important for efficient and reproducible counting, particularly in the proportional mode.

7.6.3.2 In a modification of the internal gas counter, scintillation counting has been used in place of gas-ionization counting. The inner walls of the chamber are coated with a scintillation material and the radioactive gas introduced. An optical window is made a part of the chamber, and the counting is done by placing this window on a multiplier phototube to detect the scintillations. This system is particularly useful for counting radon gas with zinc sulfide as the scintillator. Additional details on internal gas counting may be found in Reference (10).

### 7.6.4 Spectrometers and Energy-Dependent Detectors:

7.6.4.1 The availability of energy-dependent detectors (detectors whose output signal is proportional to the energy of the radiation detected) that are easy to operate and maintain and have good resolution makes it possible to measure not only the total activity of a radioactive sample but the energy spectrum of the nuclear radiations emitted. Nuclear spectrometry is most useful for alpha particles, electromagnetic radiation (gamma and X rays), and conversion electrons, since these radiations are emitted with discrete energies. Beta spectra have more limited use since beta particles are emitted from a nucleus with a continuous energy distribution up to a characteristic maximum (E-max), making a spectrum containing several different beta emitters difficult to resolve into its components. The advantages of spectrometric over total activity measurements

of radioactive sources are increased selectivity, sensitivity, and accuracy because nuclide identification is more certain, interference from other radioactive nuclides in the sample is diminished or eliminated, and counter backgrounds are reduced since only a small portion of the total energy region is used for each radiation. The detectors for alpha spectra are gridded ion-chambers and silicon semiconductor detectors. These are described in Practice D3084. A variety of semiconductors can be purchased, and these detectors have essentially replaced ion-chambers for alpha spectrometry, although the chambers have the advantages of high efficiency (nearly 50 %) for large-area sources.

7.6.4.2 The principal detectors used for gamma-ray spectrometry are high purity germanium semiconductors (HPGe) and thallium-activated sodium iodide scintillation crystals (NaI(Tl)). For X rays and very low energy gamma rays, HPGe and gas-filled thin (approximately 1 mg/cm<sup>2</sup>) window proportional counters are used. Sodium iodide is hygroscopic, so the crystal must be hermetically sealed, and the entire crystal-phototube package must be light-tight. The complete spectrometer also requires a high-voltage power supply for the phototube (usually operated at 800 to 1000 V), a preamplifier, linear amplifier, pulse-height analyzer, and output recorder. The crystal is packaged in aluminum or stainless steel. The portion of the cover through which gamma rays enter is normally thinner than the rest of the package in order to reduce low-energy photon attenuation. Sodium iodide crystals are available in a large range of sizes and shapes, from 25 by 25-mm cylinders to hemispheres and cylinders at least 305 mm in diameter. Information on the types of crystal packages and mountings that can be used is available from the manufacturers.

7.6.4.3 Germanium detectors are junction-type semiconductor devices in which a large sensitive region has been produced by the refinement of germanium to extremely low impurity levels. The crystal functions as a “solid ion chamber” when a high voltage is applied. To provide the semiconductor function, and in order to obtain optimum resolution, the detector must be operated at low temperatures to reduce thermal noise. At room temperature, sufficient free electrons will be present in the crystal to obscure the measurement of gamma and X rays (but not of alpha particles). Consequently, the HPGe detectors are operated and kept in the range of 80 to 100 degrees Kelvin by a cryostat consisting of a metallic cold-finger immersed in a dewar containing liquid nitrogen or by thermoelectric cooling. The detector is kept hermetically sealed in a vacuum to prevent impurities from condensing on the surface and lowering its resistance and is thermally and electrically isolated, usually by vacuum, to reduce heat transfer from the room to the crystal, and to maintain the high voltage required for the crystal to be a diode. A low atomic number material, such as aluminum, is the usual covering, and a molecular sieve pump is incorporated into the system to maintain the vacuum. The electronic components required to obtain spectra are similar to those for sodium iodide crystals, except that because smaller pulses must be measured, high-quality electronics are needed. The complete system includes a high-voltage bias supply for the detector (up to 5000 V for large depletion volumes), a

preamplifier (usually charge-sensitive), amplifier, biased amplifier (if needed), pulse height analyzer, and recording device. Current technology provides for digital signal processing for the signal immediately after the signal leaves the preamplifier. The digital signal processing technology provides improved resolution and peak energy stability across a large range of ambient temperatures and signal processing rates. In more advanced products, the signal processing, bias supply, and multichannel pulse height analyzer are provided as a single component.

7.6.4.4 A gamma ray entering either a NaI(Tl) crystal or a semiconductor detector may lose all or part of its energy in the detector. When all of the energy is absorbed in the detector, through multiple Compton interactions or the photoelectric effect, a full energy peak is obtained. Otherwise, only part of the energy will be observed and a Compton continuum spectrum is seen. An alternative process for high-energy gamma rays ( $>1.02$  MeV) is pair production, in which an electron-positron pair is produced, and gamma-ray peaks are observed at 0.511-MeV intervals below the full energy peak. The two most important operating characteristics of gamma detectors are efficiency and resolution. The “peak-to-Compton” or “peak-to-valley” ratio is frequently given in the literature and is related to both efficiency and resolution. These parameters should be specified by the manufacturer and the conditions under which they were measured should be given, normally by reference to an international standard (for example, ANSI N42.14) describing the measurement terms and conditions.

7.6.4.5 The resolution of a gamma-ray detector may be specified in terms of the width of the full-energy gamma-ray peak at half its maximum height—the “full width at half maximum” (FWHM)—or as the ratio of the FWHM (in units of energy) to the energy of the peak centroid. The later quantity is typically expressed as a percentage. This is shown graphically in the gamma-ray spectrum in Fig. 2. While the FWHM increases with increasing energy, the percent resolution improves. The standard for comparison is usually percent reso-

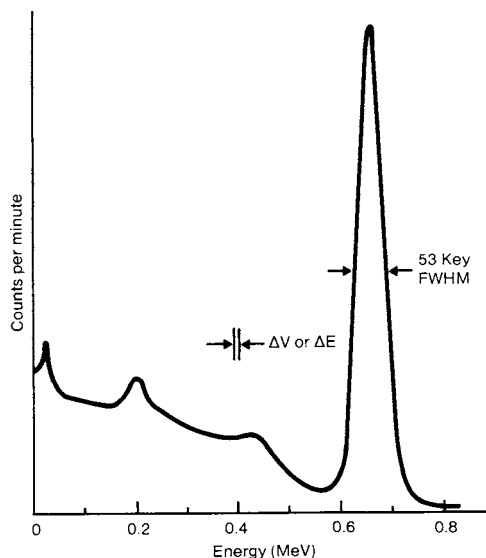


FIG. 2 Pulse Height or Energy Spectrum of Cesium-137

lution of the 0.662-MeV gamma ray emitted in the decay of  $^{137}\text{Cs}$ . Good NaI(Tl) detectors have resolutions in the range of 6.5 to 7 % for  $^{137}\text{Cs}$ . Detection efficiency for the same geometry and window thickness is a function of several parameters and much published information on efficiencies for various energies, detector sizes, source-to-detector distances, and other variables is available (11). The efficiency for gamma-ray detection may be expressed in various ways. Of primary interest in spectrometry is the full peak efficiency—the fraction of incident gamma rays that give a full-energy peak for a particular source-detector configuration. For a 102-mm thick NaI(Tl) detector, with the source on the surface (zero distance), this fraction is approximately 0.24 for the 0.662-MeV gamma-ray of  $^{137}\text{Cs}$  and approximately 0.14 for the 1.33-MeV gamma-ray of  $^{60}\text{Co}$ . The “peak-to-valley” or “peak-to-Compton” ratio is the ratio of counts at the maximum height of the full-energy peak to the counts at the minimum of the Compton continuum (Fig. 2). A high ratio indicates narrow peaks, that is, good resolution, for that particular efficiency. The Compton spectrum does not give useful information in gamma-ray spectrometry and can be considered as “noise.” The ratio varies with energy and is frequently given for the 1.33-MeV peak of  $^{60}\text{Co}$ . It increases as the crystal size increases, and, after passing through a minimum, increases as the source-to-detector distance increases, since a larger fraction of the gamma rays pass through the full depth of the crystal. A peak-to-valley ratio of 12:1 for a crystal is very good. This ratio can be increased by anti-coincidence shielding to cancel Compton events as described in 7.6.5. The efficiency of silicon for gamma-rays is considerably less than sodium iodide because of its lower atomic number (the efficiency for photoelectric absorption of gamma-rays is proportional to  $Z^5$ ) and lower density (the density of NaI is  $3.7 \text{ g/cm}^3$  and of silicon  $2.4 \text{ g/cm}^3$ ).

7.6.4.6 For a 1-MeV gamma-ray, the total absorption coefficient is about  $2 \text{ mm}^{-1}$  for sodium iodide,  $1.5 \text{ mm}^{-1}$  for silicon, and  $3 \text{ mm}^{-1}$  for germanium. As defined in IEEE and IEC standards, the efficiency of an HPGe detector is generally expressed by comparison with that of a 76 by 76-mm cylindrical NaI(Tl) detector. Comparison is made between the full-energy peak efficiencies for the 1.33-MeV gamma ray of  $^{60}\text{Co}$  when the source is 250 mm from the detector. A germanium detector with a volume of  $35 \text{ cm}^3$  has an efficiency approximately 5 % that of a 76 by 76-mm NaI(Tl) crystal. Larger HPGe detectors are now available with relative efficiencies of greater than 200 %. The very large HPGe detectors provide signal to noise ratios and resolution far superior to that offered by NaI(Tl).

7.6.4.7 There are limitations in the efficiency of the light production and collection processes in the NaI(Tl) detector system that make its resolution inferior to that of semiconductor detectors. One important factor is that about 500 eV are required to produce an electron at the photocathode in a NaI(Tl) detector system, while the average energy to produce the analogous electron-hole pair in silicon is only 3.5 eV and in HPGe 2.8 eV. The resolution of semiconductor detectors does not change greatly with energy. Presently available HPGe detectors have resolutions of 1.5 to 2.8 keV at 1.33 MeV and are from 10 to 200 % efficient as compared to a 76 by 76-mm

NaI(Tl) detector. The greater resolution makes this detector the one of choice for gamma-ray spectrometry; the ability to produce very large high purity HPGe crystals also cancels the effect of the higher efficiency previously available only from NaI(Tl). Since the pulses from a single photopeak are spread over a much smaller energy range in HPGe than in NaI(Tl), the background under the peak is much less. This means that for small sources of moderately energetic gamma-rays, HPGe is more sensitive (that is, better detection capability) than NaI(Tl). This is indicated in Table 1, where the efficiencies and backgrounds of 76 by 76-mm NaI(Tl) detector and a 35-cm<sup>3</sup>(5.5 % efficiency) HPGe detector are compared.

7.6.4.8 Spectra of beta particles and conversion electrons can be obtained with sodium iodide and semiconductor detectors sufficiently thick (a few centimetres) to absorb the particles completely. One disadvantage of NaI(Tl) and cooled semiconductors is their relatively thick entrance windows. Other semiconductor detectors, particularly the silicon surface barrier type, have thin entrance windows and can be used for beta particles at room temperature. The resolution of silicon surface barrier detectors is 5 to 10-keV for 600-keV electrons and 12 to 30-keV resolution for 5-MeV alpha particles.

7.6.4.9 Good spectra of low-energy beta particles, conversion electrons, and X rays can be obtained with a gas-flow proportional counter provided that a linear preamplifier is used. The resolution is intermediate between NaI(Tl) and HPGe. To reduce backscattering, the chamber should be made of low Z material. A counter constructed of a cylinder of graphite-impregnated plastic, poly(methyl methacrylate) ends, and a thin coaxial center wire gives good spectra for such radiations (11). A hole is cut into the outer wall and covered with aluminumized polyester film to provide a thin entrance window. Methane (10 %)–argon (90 %) is a suitable counting gas.

7.6.4.10 Organic scintillators, such as anthracene and polystyrene polymerized with scintillating compounds, are also useful for beta spectrometry. They are packaged with a phototube in a manner similar to a sodium iodide crystal. Liquid scintillation mixtures also give beta spectra, and the

output of a commercial liquid scintillation counter can be fed into a multichannel pulse-height analyzer to obtain a spectrum (2). A spectrum of <sup>210</sup>Pb <sup>210</sup>Bi <sup>210</sup>Po in Fig. 3 shows the resolution obtainable by liquid scintillation counting of aqueous samples in a dioxane-based solution. The <sup>210</sup>Bi curve is from a beta particle, and the <sup>210</sup>Po peak is from an alpha particle. Organic scintillators are preferable to NaI(Tl) for beta spectrometry because less backscattering occurs.

7.6.4.11 The output pulses of any energy-dependent detector, after linear amplification, must be sorted out according to energy to obtain the spectrum of incident radiation. The high resolution available in detectors requires analyzers with hundreds of channels to realize their full resolving power. The amplified pulse is digitized by an analog-to-digital converter (ADC), and the resulting number for a particular pulse is recorded in a pulse counter whose location is determined by digital circuitry. This makes it possible to use a digital computer to count and store in its memory the number of pulses in each channel. This conversion and storage is relatively slow, and the analyzer is blocked from processing a second pulse until the previous processing is completed. The time required to process a pulse increases with channel number. The instruments now available are sufficiently fast for almost all environmental measurement purposes. Some loss of pulse information is acceptable, as the analyzers measure and record “live time” fairly accurately. Thus, the counting time recorded by the analyzer will be the actual time it was in a condition to receive detector pulses, and not the elapsed time. To maintain good accuracy, the activity of the sample should be adjusted to give live times of 90 % or more. A computer is typically combined with the ADC and has a program to control detector operations and perform data reduction and analysis. The program may provide an output of the spectrum and results to a monitor, a printer or a digital storage device.

7.6.4.12 All multichannel pulse-height analyzers currently available are digital, and are fairly reliable instruments and relatively easy to operate. Their maintenance and repair is, however, a specialized skill similar to other computer repair. In

TABLE 1 Comparative Performance of NaI(Tl) and HPGe Gamma-Ray Detectors

Note—

NaI = 76 by 76-mm cylindrical detector.  
 Ge(Li) = 35-cm<sup>3</sup> active volume, 5.5 % efficiency.  
 A = small source placed on detector.  
 B = 57 by 57 by 57-mm thick source place on detector.  
 Counting = percent of photons emitted from the source that give a full-energy peak.  
 Shielding = 152 mm of iron, 3.2 mm of lead.  
 Counting = one 30 000-s count for both source and background.  
 time

Detection = the number of photons emitted from the source whose net limit count equals twice the counting error, or

$$N + N_B + 2(N - N_B)^{1/2}$$

where *N* is the total number of counts recorded when the source is measured and *N<sub>B</sub>* is the total number of counts recorded when the background is measured.

Photon Energy Detector	Background (cps) Under Peak	Counting Efficiency,%		Detection Limit, photons/s	
		A	B	A	B
0.14 MeV					
NaI	24	26	18	2.4	3.5
Ge	0.7	12	4	0.92	2.8
0.66 MeV					
NaI	20	14	9	4.1	6.3
Ge	0.11	1.3	0.68	3.6	6.8
1.33 MeV					
NaI	8	5.8	3.8	6.2	9.5
Ge	0.055	0.75	0.38	4.5	8.9

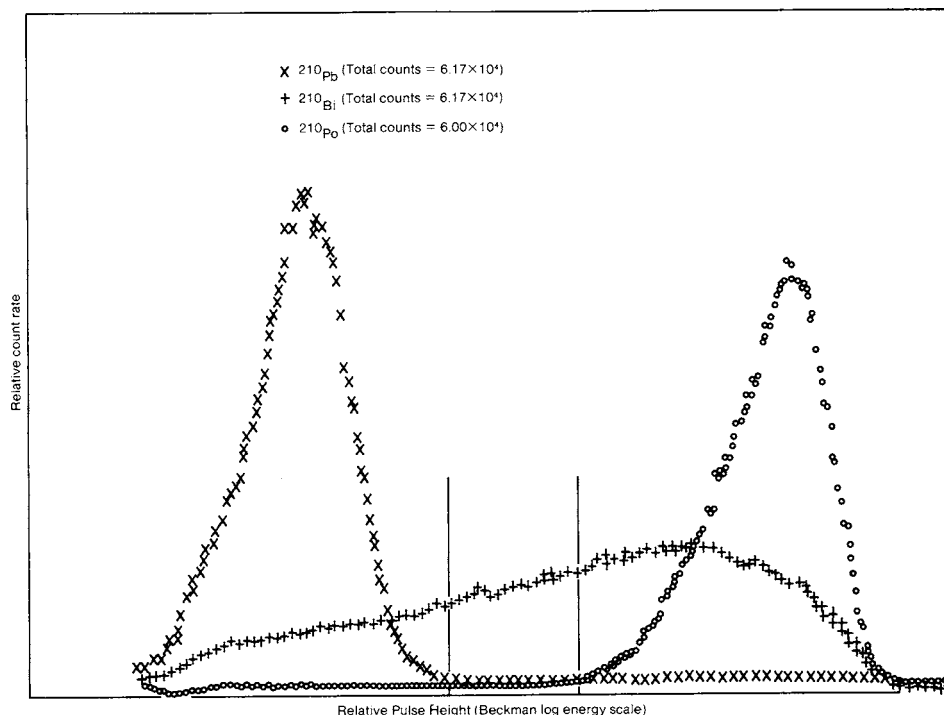


FIG. 3 Spectrum of  $^{210}\text{Pb}$   $^{210}\text{Bi}$   $^{210}\text{Po}$

comparing analyzers, some of the important specifications to consider are the number of channels, count capacity, stability, live-time accuracy, linearity, type of pulse input acceptable, and ADC speed. The minimum number of channels useful for NaI(Tl) gamma-spectrometry is 128; HPGe detectors should be used with at least a 1000-channel analyzer and alpha and beta spectra can profitably use 100 to 400 channels, depending on the energy range to be covered. Analyzers with 4096 channels are fairly common, and larger analyzers are available for special purposes.

7.6.4.13 Semiconductor detectors require low-noise, charge-sensitive amplifiers. Because of their excellent resolution, semiconductor detectors are often used with a biased amplifier following the main amplifier to isolate a portion of the spectrum for analysis. This makes it possible to use smaller analyzers than would otherwise be necessary.

#### 7.6.5 Anti-Coincidence Counters:

7.6.5.1 Substantial background reduction can be achieved in beta and gamma counters by surrounding or covering the sample detector with another detector also sensitive to beta or gamma radiation, and connecting them electronically so that any pulse appearing in both detectors is cancelled and not recorded as a count. This is usually referred to as anti-coincidence shielding, and is recommended for obtaining very low backgrounds. This type of counter was used for many years in directional studies of cosmic rays, and was first applied to reducing the background of beta counters by Libby in his study of natural  $^{14}\text{C}$ . The thick metal shielding (lead, iron, or mercury) ordinarily used to reduce cosmic-ray and gamma-ray background must also be present, and is placed outside the anti-coincidence shielding. Gas-filled beta detectors are generally shielded by gas-filled detectors, and such anti-coincidence shielding is effective primarily against the particu-

late component of cosmic rays. The anti-coincidence shielding for beta counters may consist of a number of long Geiger-Mueller tubes ("cosmic-ray counters") surrounding the sample detector or a large (approximately 152 mm square) gas-flow detector, with several anode wires so the entire area of the counter is sensitive, placed just above the sample detector. For counting solid beta sources, the sample detector has a diameter of 25 to 51 mm. Surrounding these counters on all six sides there is frequently a layer of high-purity copper to absorb gamma rays emitted from the outermost shielding, and 102 to 152 mm of lead or iron on all six sides. This is the form usually taken by the commercially available anti-coincidence shielded beta counters. Plastic or inorganic scintillators could also be used as the anti-coincidence shielding.

7.6.5.2 Anti-coincidence shielding of gamma-ray detectors operates in a similar way, and is particularly useful in reducing the Compton continuum background of gamma rays (12). Gamma rays that undergo Compton scattering and produce a pulse in both the detector and the anti-coincidence shield are cancelled electronically. Ideally, only those gamma rays that are completely absorbed in the sample detector itself produce a count that is recorded with the total energy of the gamma ray (full-energy peak). There are second-order effects that prevent complete elimination of Compton scattering, but the improvement is substantial. The anti-coincidence shield can be a large NaI(Tl) or plastic scintillator suitably attached to phototubes. They usually have a large annular hole into which the sample detector, a smaller NaI(Tl) detector, or HPGe detector is placed (13, 14).

#### 7.6.6 Coincidence Counters:

7.6.6.1 In coincidence counting, two or more radiation detectors are used together to measure the same sample, and only those nuclear events or counts that occur simultaneously



in all detectors are recorded. The coincidence counting technique finds considerable application in studying radioactive decay schemes; but in the measurement of radioactivity, the principal uses are for the standardization of radioactive sources and for counter background reduction.

7.6.6.2 Coincidence counting is a very powerful method for absolute disintegration rate measurement (6, 15). Both alpha and beta emitters can be standardized if their decay schemes are such that  $\beta$ - $\gamma$ ,  $\gamma$ - $\gamma$ ,  $\beta$ - $\beta$ ,  $\alpha$ - $\beta$ , or  $\alpha$ -X-ray coincidence occur in their decay. Gamma-gamma coincidence counting with two NaI(Tl) detectors, and the source placed between them, is an excellent method of reducing the background from Compton scattered events. Its use is limited, of course, to counting nuclides that emit two photons in cascade (which are essentially simultaneous), either directly as in  $^{60}\text{Co}$ , by annihilation of positrons as in  $^{65}\text{Zn}$ , or by immediate emission of a gamma ray following electron capture decay. If the detectors are operated with single-channel pulse-height analyzers to limit the events recorded from each detector to one of the full-energy peaks of the photons being emitted, then essentially only those photons will be counted. Non-coincident pulses of any energy in either one of the detectors will be cancelled, including cosmic-ray photons in the background and degraded or Compton scattered photons from higher energy gamma rays in the sample. Thus, the method reduces interference from other gamma emitters in the sample. If, instead of single-channel analyzers, two multichannel analyzers are used to record the complete spectrum from each detector, singly and in coincidence, then the complete coincident gamma-ray spectrum can be obtained with one measurement. The efficiency for coincidence counting is low since it is the product of the individual efficiencies in each detector, but the sensitivity is generally improved because of the large background reduction (16). This technique is often referred to as two-parameter or multidimensional gamma-ray spectrometry.

7.6.6.3 Additional background improvement is obtained if the two detectors are surrounded by a large annular NaI(Tl) or plastic scintillation detector connected in anti-coincidence with the two inner detectors. In this case a gamma ray that gives a pulse, but is not completely absorbed in one of the two inner detectors, and also gives a pulse in the surrounding detector, is cancelled electronically (13, 16). This provides additional reduction in the Compton scattering background. HPGc detectors may be used in place of the inner NaI(Tl) detectors for improved resolution and sensitivities (14).

7.7 All of the equipment described in Section 7 is available commercially.

## 8. Sampling

8.1 Collect the sample in accordance with Practice D1066 or Section 14.3 of Practices D3370.

8.2 Sample an appropriate volume depending on the expected concentration of radioactivity in the water. For precise measurements without long counting times, it is advisable to count an aliquot that contains at least 40 Bq of radioactivity.

8.3 Chemical treatment of samples to prevent biological or algal growth is not recommended and should be avoided unless essential. When necessary, select the reagents used to avoid

chemical interaction with the radioactive species in the sample. Analyze samples promptly.

8.4 Chemical treatment of samples to retain radioactive species in solution may be used but carefully select the specific treatment. The use of oxidizing acids such as  $\text{HNO}_3$  is not recommended when iodide is present since it may be oxidized to iodine and lost or be absorbed into the plastic containers if they are used. In some cases, extreme chemical treatment may be used to keep a particular chemical species in solution; examples are strongly alkaline conditions to hold molybdenum and ruthenium in solution, or acid conditions with fluoride ion to keep zirconium in solution. The addition of an acid such as hydrochloric is generally desirable to reduce hydrolysis and the loss of activity on container walls. Frequently, samples will contain insoluble material. In such cases, treat the sample by one of the following methods:

8.4.1 Filter the insoluble material and analyze both the filtrate and insoluble matter on the filter. During filtration, some material may be sorbed onto the filter and assumed to be insoluble when in fact it is soluble.

8.4.2 Centrifuge the sample and analyze both phases. Wash the insoluble phase with distilled water to remove all soluble material without dissolving the insoluble fraction.

8.4.3 In either of the above separations when the total activity is required, the insoluble matter may be dissolved and recombined with the soluble fraction. When radioactivity is left on the walls of the sample container, desorb it and add it to the sample.

8.5 Composite samples may be made by mixing aliquots of successive samples collected by an automatic sampler. Analysis of such composite samples yields average results only and loses information on short-term effects.

## 9. Instrument Operation and Control

9.1 The following procedures ensure that counting equipment is functioning properly and remains in calibration.

### 9.2 Establishing Counter Characteristics:

9.2.1 The first step in instrument control is to establish the operating characteristics of the system. Carefully measure the efficiency for counting the nuclide of interest under the conditions to be employed. Select counting conditions, that is, optimize gain, discriminator setting, and voltage for the radionuclide of interest. Set the operating voltage so that any change in counting rate is minimized for a given voltage fluctuation. Adjust the discriminators to exclude noise and unwanted interferences from the nuclides being counted. Make adjustment to optimize the signal-to-noise ratio. When the counting conditions have been selected, monitor known interferences to determine such things as the effect of counting betas in an alpha counter or alpha pulses in a beta proportional counter, etc.

9.2.2 Make daily performance checks and maintain a log for each instrument. This log should include the count for a standard and a background. When the counting rate differs statistically from the expected performance, perform additional counting to determine if the counter is malfunctioning. High background can indicate either an instrumental problem or counter contamination.



9.2.3 Certified standards are available from several suppliers. Most solution standards have the pH controlled and carrier added to ensure that hydrolysis or sorption, or both, do not change the concentration of the solution. When dilutions are made it is important to maintain the stability of the solution by diluting with a proper matrix. Store the standard in a container that minimizes evaporation by loss either through the walls or out of the stopper.

9.2.4 If a planchet is prepared as a standard, place it in a suitable container for storage, which will prevent the surface containing the activity from being contacted. A recommended practice is the preparation of two standards, using one and storing the second for periodic checks to see that the working standard has not been altered.

9.3 Counter Control and Tolerance Charts:

9.3.1 Evaluate the daily standard counts made on any counter on a statistical or tolerance basis. The best way to do this is to maintain a control chart on each counter. A control chart is a graph showing the number of counts recorded in a fixed counting period against the day of the year. Select a radioactive source having a suitable emission rate to give several thousands of counts in a relatively short counting time. Each measurement should be at least 10 000 counts in a given measurement time period. Determine the initial entry,  $\bar{C}$ , from the average of at least ten measurements over a period of days. For a statistical control chart, enter the control limit bands of  $\pm 2s_c$  and  $\pm 3s_c$  ( $s_c = \sqrt{\bar{C}}$ ) and draw lines on the chart that allow for decay of the standard over the year. For a tolerance chart, select the tolerance band for  $\bar{C}$ , based on needs, to which the counter will be held, for example, from  $\pm 1\%$  or  $\pm 3\%$  of the  $\bar{C}$ , etc. The tolerance band is equivalent to the  $\pm 3s_c$  control limit band of a control chart. Draw control chart and tolerance band lines on the chart that allow for decay of the radioactive standard over the year. An example of a control chart is shown in Fig. 4.

9.3.2 For a statistical control chart, enter the result of the standard count in the control chart and take the following action:

9.3.2.1 If the result is inside the  $\pm 2s_c$  band, consider the counter to be in control.

9.3.2.2 If the result lies outside the  $\pm 2s_c$  band, but inside the  $\pm 3s_c$  band, consider the counter to be in control but flag this result.

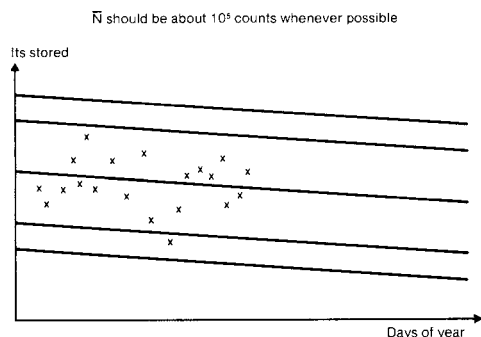


FIG. 4 Typical Counter Control Chart

9.3.2.3 If the result lies outside the  $\pm 3s_c$  band, consider the counter out of control. Corrective action is needed if repeated counts remain outside the  $\pm 3s_c$  band.

9.3.3 For a tolerance chart, enter the result of the standard count in the tolerance chart and take the following action:

9.3.3.1 If the result lies outside the tolerance band, consider the counter out of control. Corrective action is needed if repeated counts remain outside the tolerance band.

9.3.4 For alpha and gamma spectrometry, it is also important to monitor for system resolution (see Fig. 5).

9.3.5 In addition to the control charts made on each counter, keep all pertinent information about the system in a log book or permanent file.

Counter Logs

$\alpha$ and $\beta$ Counters	$\gamma$ Spectrometers
standard counts	standard counts
background counts	standard resolution
system changes	background counts
control charts	control charts

10. Counting Statistics

10.1 Each nuclear disintegration (6, 17, 18) is a completely random and independent process. Established methods of statistical analysis are available to describe the random phenomenon of nuclear disintegration. The total number of particles counted in a time period can be shown to deviate from the average in accordance with the expression,  $C \pm \sqrt{C}$  where  $C$  is the number of counts in the counting period  $t$ . Similar behavior is found in the background count rate for any counter and the recorded background will deviate from the average in accordance with the expression  $C_b \pm \sqrt{C_b}$  where  $C_b$  is the number of counts recorded in the counting period  $t_b$ . In the situation where the net count is 0, the  $\sqrt{N}$  expression may be replaced by  $\sqrt{N + 1}$ . Determine the net counting rate as follows:

$$C_n = \text{Net count rate} = \left( \frac{C}{t} - \frac{C_b}{t_b} \right) \tag{1}$$

The standard deviation (uncertainty at the 68.3 % confidence level) of the net count rate,  $C_n$ , is defined as follows:

$$s_n = \sqrt{(C/t)^2 + (C_b/t_b^2)} \tag{2}$$

or for the net count rate with its standard deviation:

$$\text{Net count rate} = \left( \frac{C}{t} - \frac{C_b}{t_b} \right) \pm \sqrt{\frac{C}{t^2} + \frac{C_b}{t_b^2}} \tag{3}$$

In practice, many counts will be made that deviate from the average by more or less than the standard deviation, since  $s_n$  by definition would give those limits  $X - s_n$  and  $X + s_n$ , which

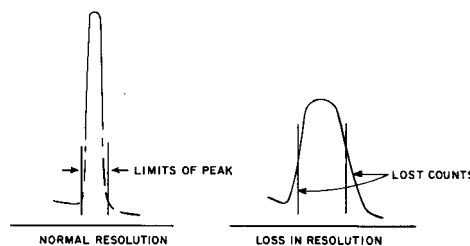


FIG. 5 System Resolution in Alpha and Gamma Spectrometry

would include approximately 68 % of all observed values. In terms of the population of observations between limits, one can express the chance or probability of a result being between the limits  $X - ks_n$  and  $X + ks_n$  as “confidence levels,” where  $k$  is simply a multiplier for  $s_n$ . It may also be described as the “coverage factor” as used in the GUM (see ISO Guide to the Expression of Uncertainty in Measurement). In evaluating the effect of  $k$  for total counts (per measurement period) greater than  $\sim 60$ , one can tabulate  $k$  and the “confidence level.”

Confidence Level, %	$k$
68.3	1.0
90	1.65
95	1.96
99	2.58
99.7	3.0

10.2 The 95 % confidence limit is frequently used and has been generally accepted since counts outside this band are considered statistically significant. In practice,  $k$  is often rounded to 2.0 and the term  $2s_n$ , or  $2\sigma$ , standard deviation is often used. In some cases the  $3s_n$ , or  $3\sigma$ , standard deviation is used and gives the 99.7 % confidence limits.

10.3 Precision—There is a measure of the reproducibility of a measurement. There are a number of items that affect the precision of radioactivity measurements, the more important of these are as follows:

10.3.1 Position—For point sources the observed radioactivity is proportional to the reciprocal of the square of the distance of the source from the detector. Measure all sources being directly compared at the same source-to-detector distance.

10.3.2 Radiation Scattering—Changes in the sample support and backing material can seriously affect the precision of radiation measurements, particularly that of beta radiation. Standards and samples should have the same backing material. The use of a sample support rack constructed of a material with low atomic number will reduce the effect of side-scattering. Scattering from backing material can be minimized by the use of a thin, low atomic number material.

10.3.3 Background—Measure this radioactivity with no sample near the detector and subtract from all measurements of gross sample activity. This requirement becomes more critical as the net sample activity becomes small with respect to the background. Perform routine periodic measurements of the background to check for possible detector contamination or malfunction.

10.3.4 Absorption—Alpha and beta radiations are partly absorbed by the sample and by all substances separating the sample from the detector. This effect is usually small for gamma radiation but beta and alpha radiation are seriously affected. If sources of the same atomic number and mass are compared on the same counter in the same geometry, absorption will be constant under these conditions but should not be ignored. Since the sample mass may vary significantly, construct calibration curves to correct for changes in self-absorption.

10.3.5 Quenching—This is any process that reduces the photon output in a scintillation system; this in turn reduces the measured count rate. Quenching can be caused by such things as sample color and chemicals in the sample. The need to correct for this phenomenon can be avoided if samples of the

same color and same chemical composition are compared. If this is not possible, most instrument manufacturers and texts describe methods for construction of calibration curves to correct for this phenomenon (19, 20, 21).

10.4 Overall Uncertainty in a Determination—Report measurement results with the estimated overall measurement uncertainties shown. This overall measurement uncertainty may be expressed as the relative standard deviation (RSD). There are two approaches to this: (1) rigid propagation of uncertainties, which is not sound in practice since the individual components are not well characterized, and (2) a combination of known uncertainties. In combining uncertainties, use the following relationship:

$$U = \sqrt{U_1^2 + U_2^2} \tag{4}$$

where:

- $U$  = overall uncertainty of the measurement (RSD),
- $U_1$  = random counting uncertainty (RSD), and
- $U_2$  = other random uncertainties (RSD).

The work can be divided into two main classes, as follows:

10.4.1 Gamma Spectrometry—Assuming that the integrity of the sample is known, the sampling and treatment effects are at a minimum and then only two significant terms are present. These are accuracy of calibration and precision of counting:

$$U_{\gamma spec} = \sqrt{U_c^2 + U_a^2} \tag{5}$$

where:

- $U_{\gamma spec}$  = overall error for the measurement (RSD),
- $U_c$  = calibration uncertainty (RSD), and
- $U_a$  = counting uncertainty (RSD).

10.4.2 Separative Work—The sample treatment introduces uncertainty into the measurement. One estimate of great merit is as follows:

$$U = \sqrt{U_m^2 + U_a^2 + U_c^2} \tag{6}$$

where:

- $U$  = overall uncertainty for the measurement,
- $U_m$  = intrinsic precision of the method,
- $U_a$  = counting uncertainty, and
- $U_c$  = calibration uncertainty.

The intrinsic precision may be determined by doing a single-operator precision (SOP) test at three or four activity levels.

Level, dps	SOP Test Data			Average } $S_M$
	$S_x$ (%RSD)	$S_d$ (%RSD)	$S_M$ (%RSD)	
0.2	8	$S_{d1}$	$S_{M1}$	}
2	4	$S_{d2}$	$S_{M2}$	
200	2	$S_{d3}$	$S_{M3}$	

The use of methods that have been round-robin tested will provide a method where  $s_n$  is known.

10.4.3 The overall uncertainty of a measurement may be used to make a statistical comparison between two duplicate or replicate results. This can be done using a relationship of following form (39):

$$\frac{A_{a1} - A_{a2}}{\sqrt{S_{x1}^2 + S_{x2}^2}} \tag{7}$$

The resulting value of this relationship can be used to assess the statistical agreement between the two results.