

15 NUCLEAR COUNTING INSTRUMENTATION

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15.1 Introduction

This chapter presents descriptions of counting techniques to help the user to determine what radioanalytical measurement method(s) best suit a given need. References cited in the text provide additional details of how these measurements are made. The primary focus here is on the variables that ultimately affect the bias and precision of the counting data. The type of information that is desired—in relation to the type of radiation to detect—will determine the type of instrument and associated technique one will use to generate data. For example, samples containing a single radionuclide of high purity, sufficient energy, and ample activity may only require a simple detector system. In this case, the associated investigation techniques may offer no complications other than those related to calibration and reproducibility. At the other extreme, a sample or set of samples may require quantitative identification of many radionuclides or the laboratory may need to prepare unique calibration standards. In the latter case, specialized instruments are available. Typically, a radiochemical laboratory will encounter samples routinely that require a level of information between the two extremes described above.

A typical laboratory may be equipped with the following nuclear counting instrumentation:

- Proportional or Geiger-Mueller detectors for alpha and beta counting;
- Sodium iodide or high resolution germanium detectors for gamma detection and spectrometry;
- Solid state detectors for alpha spectrometry;
- Scintillation counters suitable for both alpha- or beta-emitting radionuclides; and
- Multichannel analyzers for alpha and gamma-ray spectrometry.

A basic requirement for accurate measurements is the use of high quality standards, traceable to a national standards organization (Section 15.9; ANSI N42.22, ANSI N42.23), to calibrate

30 instrumentation. Generally, with the present availability of good standards, radiochemistry
31 laboratories rarely require instrumentation suitable for producing their own calibration standards.
32 However, it is always advisable to compare each new standard received against the previous
33 standard. The next three main sections of the chapter describe counting instrumentation for alpha
34 (Section 15.2), beta (Section 15.3), and gamma (Section 15.4) radiation. In a number of cases the
35 same instrumentation is used for radionuclides with one or more types of radiation. Note that a
36 review covering descriptions of radionuclides, types of radiation, associated principles, and
37 definitions for related terminology is given in Appendix A of this manual. The discussion next
38 turns to several specific areas to cover spectrometry (Section 15.5), special instrumentation
39 (Section 15.6), and spectrometers and energy-dependent detectors (Section 15.7). Shielding
40 (Section 15.8) to reduce detector background of nuclear counting instruments and instrument
41 calibration (Section 15.9) follows. This chapter closes with a discussion of other nuclear
42 counting instrumentation considerations (Section 15.10) including a discussion on non-nuclear
43 instrumentation (Section 15.10.4).

44 **15.2 Alpha Counting**

45 **15.2.1 Introduction**

46 Alpha particles are relatively massive, expend their energy over short distances, and typically
47 exhibit limited penetration into neighboring materials. Alpha particles are also characterized by
48 an intense loss of energy while passing through matter (see ICRU, 1992, for a discussion of dose
49 equivalents and linear energy transfer). This loss of energy is used to differentiate alpha
50 radioactivity from other types through the dense ionization or intense scintillation it produces.
51 This high rate of loss of energy in passing through matter, however, also makes sample
52 preparation conditions for alpha counting more stringent than is necessary for other types of
53 radiation. An example of direct alpha counting to determine total alpha activity is given in
54 ASTM C799.

55 Alpha radioactivity normally is measured by one of several types of detectors in combination
56 with suitable electronic components. The detector devices most used are ionization chambers,
57 proportional counters, silicon semiconductor detectors, and scintillation counters. The associated
58 electronic components in all cases include high-voltage power supplies, preamplifiers, amplifiers,
59 scalars, analog-to-digital converters, and recording devices.

60 The measured alpha-counting rate from a sample will depend on a number of variables. The most
61 important of these variables are:

- 62 • Geometry;
- 63 • Source diameter;
- 64 • Self-absorption;
- 65 • Absorption in air and detector window;
- 66 • Coincidence losses; and
- 67 • Backscatter.

68 These are discussed in detail in the literature (Blanchard et al., 1960; Hallden and Fisenne, 1963),
69 and can be measured or corrected for in many cases by holding conditions constant during the
70 counting of samples and standards.

71 Alpha counters have low backgrounds and high efficiencies. Thus, outside sources of alpha
72 radiation will not impact the counting process and the instrument essentially focuses on the alpha
73 source presented by the sample. However, some counters are easily contaminated internally and
74 care should be taken to avoid contamination. Silicon detectors operated in a vacuum may become
75 contaminated due to recoil from sources (Merritt et al., 1956). Some alpha counters are sensitive
76 to beta radiation depending on the detector (Blanchard et al., 1960; Hallden and Fisenne, 1963).
77 In these cases, electronic discrimination is often used to eliminate the smaller pulses due to beta
78 particles. A discussion of alpha particle attenuation can be found in Section 15.10.1.1.

79 **15.2.2 Detectors for Alpha Counting**

80 15.2.2.1 Ionization Chambers

81 As the incident particle enters the ionization chamber, ionization occurs through the interaction
82 of the particle with the fill gas. The secondary electrons produced through these interactions are
83 accelerated toward the anode as a result of the bias applied to the system. An ion current is
84 produced at the anode as a result of the collection of the free electrons (negative ions) generated
85 through ionization interactions. The charge collected at the anode is collected across an RC
86 circuit resulting in a change in potential across a capacitor. The change in potential is thus related
87 to the charge produced from the collection of electrons produced through the ionization
88 interactions of the incident particle.

89 15.2.2.2 Proportional Counters

90 As the incident particle enters the proportional counter, ionization occurs through the interaction
91 of the particle with the fill gas. The secondary electrons produced through these interactions are
92 accelerated toward the anode as a result of the bias applied to the system. In proportional

93 counters, the free electrons gain sufficient kinetic energy during acceleration to produce
94 secondary ionization as they migrate toward the anode. This effect, known as “gas multiplica-
95 tion,” is used to amplify the charge collected at the anode. Similar to ionization chambers, the
96 charge collected at the anode is collected across an RC circuit resulting in a change in potential
97 across a capacitor. As a result of gas multiplication, the voltage pulse produced is considerably
98 larger than the pulse produced in an ionization chamber. The magnitude of the voltage pulse is
99 thus proportional to the original number of ion pairs formed by the incident particle.

100 Proportional detectors are generally constructed of stainless steel, oxygen free/high conductivity
101 (OFHC) copper, or aluminum. No additional shielding is required for alpha proportional
102 counting. The counter should be capable of accepting mounts up to 51 mm in diameter.
103 Proportional counters are available in two types, either with or without a window between the
104 sample and the counting chamber. The manufacturer’s specifications for either type should
105 include performance estimates of background count rate, length and slope of the voltage plateau,
106 and efficiency of counting a specified electrodeposited standard source, along with the type of
107 gas used in the tests. For a window flow counter, the window thickness—in milligrams per
108 square centimeter—also should be specified. With a windowless flow counter the sample and
109 sample mount should be made of an electrical conductor in order to avoid erratic behavior due to
110 static charge buildup.

111 Typical parameters for the alpha windowless flow counter are:

112 background count rate = 10 counts/h or 2.8×10^{-3} cps
113 length of voltage plateau = 300 V
114 slope of voltage plateau = 1%/100 V for an electrodeposited source

115 For a window flow counter, typical values are:

116 window thickness = 0.08 to 0.5 mg/cm²
117 background count rate = 10 counts/h or 2.8×10^{-3} cps
118 length of voltage plateau = 300 V
119 slope of voltage plateau = 1%/100 V for an electrodeposited source
120 efficiency = 35 to 40 percent for an electrodeposited source

121 15.2.2.3 Scintillation Counters

122 In a scintillation counter, the alpha particle transfers energy to a scintillator, such as zinc sulfide
123 (silver activated). The transfer of energy to the scintillator results in the production of light at a

124 wavelength characteristic to the scintillator, and with an intensity proportional to the energy
125 transmitted from the alpha particle. The scintillator medium is placed in close proximity to the
126 cathode of a multiplier phototube; light photons from the scintillator strike the photo cathode,
127 and electrons are emitted. The photoelectrons are passed through a series of dynodes resulting in
128 the multiplication of electrons at each stage of the multiplier phototube. After amplification, a
129 typical scintillation vent will give rise to 10^7 to 10^{10} electrons, which is sufficient to serve as a
130 signal charge for the scintillation event. The electrons are collected across an RC circuit, which
131 results in a change in potential across a capacitor, thus giving rise to a pulse used as the
132 electronic signal of the initial scintillation event.

133 The counter size is limited by the multiplier phototube size, a diameter of 51 mm being the most
134 common. Two types of systems may be employed. In the first, the phosphor is optically coupled
135 to the multiplier phototube and either is covered with a thin ($<1 \text{ mg/cm}^2$) opaque window or
136 enclosed in a light-proof sample changer. With the sample placed as close as possible to the
137 scintillator, efficiencies approaching 40 percent may be obtained. The second system employs a
138 bare multiplier phototube housed in a light-proof assembly. The sample is mounted in contact
139 with a disposable zinc sulfide disk and placed on the phototube for counting. This system gives
140 efficiencies approaching 50 percent, is associated with a slightly lower background, and less
141 chance of counter contamination.

142 A major advantage of alpha scintillation counting is that the sample need not be conducting. For
143 a 51 mm multiplier phototube with the phosphor coupled to the tube, typical values obtained are
144 a background count rate of 0.006 cps and an efficiency for an electrodeposited standard source of
145 35 to 40 percent. With a disposable phosphor mounted on the sample, typical values are a
146 background count rate of 0.003 cps and an efficiency for an electrodeposited standard source of
147 45 to 50 percent. For both systems, voltage plateau length is 150 V with a slope of 5
148 percent/100 V.

149 15.2.2.4 Liquid Scintillation Counters

150 Liquid scintillation counting of alpha emitters with a commercially available instrument
151 overcomes many of the problems inherent in other techniques (Passo and Cook 1994; Horrocks,
152 1974; DeFilippis, 1990; Friedlander et al., 1964; Curtis et al., 1955; Matt and Ramsden, 1964;
153 Overman and Clark, 1960; Price, 1964; Flynn et al., 1971). Typical background counting rates
154 range from 0.1 to 0.2 cps. Sample preparation, after radiochemical separation is performed,
155 involves mixing the sample aliquant with a suitable liquid scintillator solution or gel phosphor
156 before counting. In this way, planchet preparation is eliminated, volatile components are retained,
157 and the completely enclosed sample cannot contaminate the counting chamber. Ideally, the

158 sample is uniformly distributed in the scintillator so there is no self-absorption. This results in a
159 counting efficiency of almost 100 percent. Because of the high alpha energies, considerable
160 chemical quenching effects can be tolerated before counting efficiency is reduced. Coincidence
161 losses are small in liquid scintillation counting at count rates up to 2×10^4 cps. For samples that
162 contain both alpha and high-energy beta emitters, difficulties do arise in distinguishing between
163 the two. The problem is due primarily to the broad continuum of beta energy distribution up to
164 the maximum energy and the poor resolution of liquid scintillation spectrometers. This problem
165 is aggravated because the light yield per million electron volts of alpha particles in most liquid
166 scintillators is approximately tenfold lower than a beta particle of equivalent energy, putting the
167 pulses from alphas and high-energy betas in the same region. Correction for beta activity may be
168 made by certain mathematical, graphical or electronic techniques (see discussion of pulse shape
169 discrimination in Section 15.5.4). It is preferable to separate the alpha emitter from the bulk of
170 the beta activity by chemistry.

171 15.2.2.5 Semiconductor Detectors

172 Semiconductor detectors used for alpha counting are essentially solid-state ionization chambers.
173 The ionization of the gas in an ionization chamber by alpha particles produces electron-ion pairs,
174 while in a semiconductor detector electron-hole pairs are produced. The liberated charge is
175 collected by an electric field and amplified by a charge-sensitive amplifier. In general, ion-
176 implanted-silicon or silicon surface barrier detectors are used for alpha counting. These detectors
177 are n-type base material upon which gold is evaporated to make a contact. The semiconductor
178 material must have a high resistivity since the background is a function of the leakage current.
179 This leakage current is present in an electric field since the starting material is a semiconductor,
180 not an insulator. The leakage current of silicon diodes doubles for every 5.5 to 7.5 °C change
181 in ambient temperature. Since the preamp HV bias resistor is a noise contributor, it is necessarily
182 of high value, typically 100 megohm. With a surface barrier detector having leakage current of
183 0.5 μ A, the change in bias voltage at the detector for a 2 °C change in ambient temperature can
184 be as much as 13V. This is enough bias change to affect overall gain of the detector-preamplifier
185 by a substantial amount. The reversed bias that is applied reduces the leakage current and a
186 depletion layer of free-charge carriers is created. This layer is very thin and the leakage current is
187 extremely low; therefore, the interactions of photons with the detector will have negligible effect.
188 Since the detector shows a linear response with energy, any interactions of beta particles with the
189 detector can be eliminated by electronic discrimination. The semiconductor is of special interest
190 in alpha counting where spectrometric measurements may be made since the average energy
191 required to produce an electron-hole pair in silicon is 3.5 ± 0.1 eV compared to the 25 to 30 eV
192 needed to produce an ion pair in a gridded ionization chamber. Consequently, silicon detectors
193 provide much improved resolution and also normally have lower background count rates.

194 The detector size is generally less than 25 mm in diameter since the resolution decreases and cost
195 increases with detector size. For best results, the sample should be electrodeposited to make a
196 lower mass source (Puphal and Olson, 1972). However, micro precipitation as fluorides has been
197 reported with only slight lose of resolution (Sill and Williams, 1981; Hindman, 1983). The
198 detector is operated in a vacuum chamber. Typical backgrounds range from 8×10^{-5} to 2×10^{-4} cps.

199 **15.3 Beta Counting**

200 **15.3.1 Introduction**

201 This section covers the general techniques used to measure the beta particle activity resulting
202 from radiochemical separations of specific nuclides or groups of nuclides. Beta radioactivity may
203 be measured by several types of instruments that provide a detector and a combined amplifier,
204 power supply, and scaler. The most widely used detectors are proportional or Geiger-Mueller
205 counters—however, scintillation systems offer certain advantages (see discussion in Section
206 15.3.3). An example of the measurement of fission product activity by beta counting is given in
207 ASTM C799, D1890, and D3648.

208 **15.3.2 Proportional Counter**

209 Among the gas ionization-type detectors, the proportional type counter is preferable because of
210 the shorter resolving time and greater stability of the instrument. For preparing solid sources for
211 beta activity measurement, the sample is reduced to the minimum weight of solid material having
212 measurable beta activity by dissolution, radiochemistry, precipitation, or ion exchange tech-
213 niques. For measuring solid sources resulting from individual radiochemical separation
214 procedures, the precipitate is appropriately mounted for counting.

215 Beta particles entering the sensitive region of the detector produce ionization that is converted
216 into an electrical pulse suitable for counting. The number of pulses per unit time is directly
217 related to the disintegration rate of the sample by an overall efficiency factor. This factor
218 combines the effects of sample-to-detector geometry, sample self-shielding, backscatter,
219 absorption in air and in the detector window (if any), and detector efficiency. Because most of
220 these individual components in the overall beta-particle detection efficiency factor vary with beta
221 energy, the situation can become complex when a mixture of beta emitters is present in the
222 sample. The overall detection efficiency factor may be empirically determined with prepared
223 standards of composition identical to those of the sample specimen, or an arbitrary efficiency
224 factor can be defined in terms of a single standard such as cesium-137 (^{137}Cs) or other nuclide.

225 Gross counts can provide only a very limited amount of information and therefore should be used
226 only for screening purposes or to indicate trends.

227 **15.3.3 Liquid Scintillation**

228 Liquid scintillation counting (LSC) avoids many sources of error associated with counting solid
229 beta sources, such as self-absorption, back scattering, loss of activity during evaporation due to
230 volatilization or spattering, and variable detection efficiency over a wide beta-energy range. In
231 addition to the greatly improved accuracy offered by liquid scintillation counting, sample
232 preparation time and counting times are significantly shorter. Sample preparation involves only
233 adding a sample aliquant to the scintillator or gel phosphor. Because every radioactive atom is
234 essentially surrounded by detector molecules, the probability of detection is quite high even for
235 low-energy beta particles. Radionuclides having maximum beta energies of 200 keV or more are
236 detected with essentially 100 percent efficiency. Liquid scintillation can, at times, be disadvan-
237 tageous due to chemiluminescence, phosphorescence, quenching, or the typically higher
238 backgrounds.

239 The observed count rate for a liquid scintillation sample is directly related to the beta (plus
240 conversion electron) and positron emission rate in most cases. The important exceptions are: beta
241 emitters whose maximum energy is below 200 keV, and counting systems wherein quenching
242 decreases the expected photon yield, thereby decreasing the overall detection efficiency
243 significantly below 100 percent. Low-energy beta emitters, such as tritium (^3H) or carbon-14
244 (^{14}C), can be measured accurately only when the appropriate detection efficiency factor has been
245 determined with a known amount of the same radionuclide counted under identical conditions.
246 Quenching losses are greatest at low beta energies. Quenching may be evaluated by comparison
247 to known quench standards of the same radionuclide, using the channel ratio technique, or with
248 other techniques as described in the manufacturer's instructions.

249 For measurements in which data are expressed relative to a defined standard, the individual
250 correction factors cancel whenever sample composition, sample weight, and counting
251 configuration and geometry remain constant during the standardization and tests.

252 Liquid scintillation counting systems use an organic phosphor as the primary detector. This
253 organic phosphor is combined with the sample in an appropriate solvent that achieves a uniform
254 dispersion. A second organic phosphor often is included in the liquid scintillation cocktail as a
255 wavelength shifter. The wave length shifter efficiently absorbs the photons of the primary
256 phosphor and re-emits them at a longer wavelength more compatible with the multiplier
257 phototube. Liquid scintillation counting systems use either a single multiplier phototube or two

258 multiplier phototubes in coincidence. The coincidence counting arrangement is less likely to
259 accept a spurious noise pulse that occurs in a single phototube, and thus provides lower
260 background. The requirement that both multiplier phototubes respond to each has a slight effect
261 on the overall detection efficiency of betas with E-max >200 keV; however, system response to
262 beta E-max <200 keV will be significant. The need to minimize detectable radioactivity in the
263 detector and its surroundings is likewise important in liquid scintillation counting. To achieve
264 this, scintillation-grade organic phosphors and solvents are prepared from low ¹⁴C materials such
265 as petroleum. The counting vials are of low potassium glass or plastic to minimize counts due to
266 potassium-40 (⁴⁰K). Liquid scintillation provides a fixed geometry from a given size counting
267 vial and liquid volume. The calibration of liquid scintillation counting detectors is given in
268 ASTM E181. The use of an organic phosphor for liquid scintillation counting creates a mixed
269 waste. Chapter 20 of this manual addresses the proper disposal of these materials.

270 Another approach to LSC without the use of organic phosphors is Cerenkov counting. When
271 charged particles pass through a dielectric medium, such as water, and there is an exchange of
272 energy to the molecules of that medium, Cerenkov radiation is produced. This happens if the
273 charged particles are moving faster than the speed of light and the exchange of energy produces
274 electronic polarization, then when the polarized molecules return to a normal state the excess
275 energy is released as electromagnetic radiation (Kessler, 1986). Wave shifters are usually
276 employed to convert the ultraviolet Cerenkov radiation to the visible range. Although Cerenkov
277 counting efficiencies are about 20 to 50 percent (Scarpitta and Fisenne, 1996) lower than when
278 organic phosphors are used, mixed waste disposal is eliminated.

279 **15.3.4 Solid Organic Scintillators**

280 Organic scintillators, such as p-terphenyl plus a wave shifter in a plastic monomer, are
281 polymerized to form sheet material of any desired thickness. The plastic phosphor counting
282 system (Campion et al., 1960) has its widest use as a beta particle detector for separated, solid
283 samples rather than for beta spectrometry applications.

284 The plastic beta scintillator phosphor is mounted directly on the sample and is discarded after
285 counting. The phosphor-sample sandwich is placed in direct contact with the multiplier
286 phototube yielding essentially a 2- π configuration. Since the output pulse of the detector system
287 is energy dependent, the counting efficiency for a given phosphor thickness of 0.25 mm yields
288 the highest counting efficiency with the lowest background.

289 Solid samples (precipitates from radiochemical separations) containing 3 to 5 mg/cm² of stable
290 carrier are measured in such a system. For yttrium-90 (⁹⁰Y) a solid sample of this type would
291 have a counting efficiency of 45 to 50 percent.

292 A plastic scintillator/phosphor system with a 25 mm multiplier phototube shielded with 12.7 mm
293 of lead has background in the order of 4×10^{-2} cps. For very low backgrounds, about 4×10^{-3} cps,
294 the multiplier phototube and sample assembly are fitted into a well-type hollow anode Geiger
295 tube operated in anti-coincidence. The entire assembly is then placed in a heavy shield.

296 The system has many advantages but reduction of background is probably most important. The
297 reduction occurs since the scintillator does not see the surrounding mechanical components of
298 the counter. The additional advantage of keeping the counter itself free from contamination by
299 enclosing the phosphor-sample sandwich is also important.

300 A note of caution is advisable at this point. Any beta particle detection system, whether internal
301 gas counters or scintillation counters, will detect alpha particles. It is not possible to
302 electronically discriminate against all the alpha pulses.

303 If a sample is suspected of containing alpha activity, a separate alpha measurement should be
304 made to determine the alpha contribution to the beta measurement.

305 **15.3.5 Beta Particle Counter**

306 The end-window Geiger-Mueller tube and the internal proportional gas-flow chambers are the
307 two most prevalent types of detectors. Other types of detectors include scintillators and solid-
308 state detectors. The material used in the construction of the detector and its surroundings should
309 contain a minimal level of detectable radioactivity. If the detector is of the window-type, the
310 window thickness may be used in calculating beta-ray attenuation; however, direct calibration of
311 the entire counting system with standards is recommended. The manufacturer should provide all
312 settings and data required for reliable and accurate operation of the instrument. Detectors
313 requiring external positioning of the test sample should include a support of low-density material
314 (aluminum or plastic), which ensures a reproducible geometry between the sample and the
315 detector. Because different sample to detector geometries are convenient for differing sample
316 activity levels, the sample support may provide several fixed positions ranging from 5 to 100 mm
317 from the detector.

318 The detection capability for both Geiger-Mueller and proportional counters is a function of the
319 background counting rate. Massive shielding or anti-coincidence detectors and circuitry, or both,

320 are generally used to reduce the background counting rate to increase the lower limit of detection
321 (Friedlander et al., 1964). ASTM E181 covers the procedure for the calibration of beta particle
322 counting detectors. An application of beta particle counting is given in ASTM E1005.

323 **15.3.6 Associated Electronic Equipment**

324 The high voltage power supply amplifier, scaler, and mechanical register normally are contained
325 in a single chassis. The power supply and amplifier sections are matched with the type of detector
326 to produce satisfactory operating characteristics and to provide sufficient range in adjustments to
327 maintain stable conditions. The scaler should have a capacity for storing and visually displaying
328 at least 9×10^5 counts. The instrument should have an adjustable input sensitivity matched to that
329 of the detector, and variable high voltage power supply—an adjustable power supply and meter
330 are unnecessary for liquid scintillation systems. Counting chambers of Geiger-Mueller and
331 proportional counters contain a suitable counting gas and an electrode. Counting rates that
332 exceed 200 cps should be corrected for dead time loss when using a Geiger-Mueller tube. As the
333 applied voltage to the electrode is increased, the counting chamber exhibits responses that are
334 characteristic of a particular voltage region. At low voltages of the order of 100 V, there is no
335 multiplication of the ionization caused by a charged particle. At voltages approaching 1,000 V,
336 there is appreciable amplification of any ionization within the counting chamber; however, the
337 size of the output pulse is proportional to the amount of initial ionization. When operated in this
338 voltage region, the device is known as a proportional counter. Usually, there is a region at least
339 100 V wide, known as a plateau, wherein the count rate of a standard is relatively unaffected. The
340 operating voltage for proportional counters is selected to approximate the middle of this plateau
341 in order to maintain stable responses during small voltage shifts. The plateau region is
342 determined by counting a given source at voltage settings that differ by 25 or 50 V. The number
343 of counts at each setting is recorded, and the resultant counts versus voltage are plotted. Voltage
344 plateau curves are to be re-measured periodically to ensure continued instrument stability, or
345 whenever an instrument malfunction is indicated. If the voltage is increased beyond the
346 proportional region into the 1,500 to 2,000 V region, the pulse size increases and the dependence
347 on the initial ionization intensity disappears. This is the beginning of the Geiger counting region,
348 where a single ion pair produces the same large pulse as an intense initial ionization.

349 In order to eliminate alpha particle interferences a thin absorber between the sample source and
350 the detector can be used. The absorber diameter should exceed that of the detector window. The
351 absorber should be placed against the window to minimize beta particle scatter. Any absorber
352 that stops alpha particles will also attenuate low energy beta particles somewhat. For example, an
353 aluminum absorber of 7 mg/cm^2 will absorb 48 percent of beta particles of 350 keV maximum
354 energy. Chemical separation of the alpha and beta particle emitters produces a higher degree of

355 accuracy for internal detector measurements. Published information on beta particle absorption
356 (Friedlander et al., 1964) should be used as a guide for use of an absorber. In liquid scintillation
357 spectra, the alpha component appears as a peak on the beta continuum and thus provides a basis
358 for resolving the two (Bogen and Welford, 1971).

359 **15.4 Gamma Counting**

360 **15.4.1 Introduction**

361 This section covers the non-destructive measurement of gamma-ray radioactivity. Since gamma
362 radiation is a penetrating form of radiation, it can be used for non destructive measurements of
363 samples of any form and geometry as long as standards of the same form are available and are
364 counted in the same geometry to calibrate the detector. Because of this penetrating nature,
365 attenuation, because of variations in sample density or sample thickness, although usually not
366 significant, can be mathematically corrected.

367 When a standard cannot be obtained in
368 the matrix and density of samples being
369 counted, a correction for the different
370 absorption in the matrices should be
371 made (Modupe et al., 1993). Photons
372 interact with matter in one of three
373 ways: photoelectric, where all energy is
374 transferred; Compton scattering, where
375 only part of the energy is transferred;
376 and pair production, where the energy
377 creates a positron-electron pair. When
378 the positron annihilates the electron, two
379 511 keV photons are emitted. Figure
380 15.1 shows the relative probability of
381 each of the three predominant photon
382 interactions with germanium.

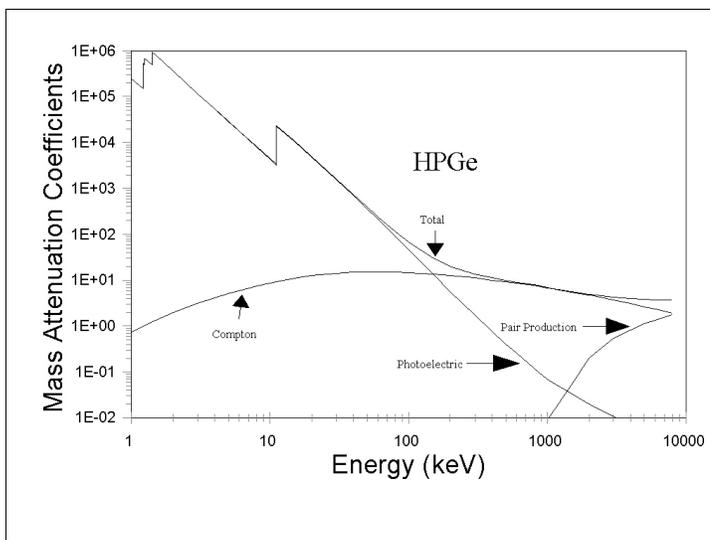


FIGURE 15.1 Gamma-ray Interactions with Germanium

383 Since different nuclides emit distinct and constant spectra of gamma radiation, the use of an
384 energy discriminating system provides identification and measurement of all the components
385 present in a mixture of radionuclides. General information on gamma-ray detectors and gamma
386 counting is covered in the literature (Friedlander et al., 1964, and ICRU, 1994). Recent applica-
387 tions of gamma counting are given in several ASTM Test Methods (ASTM C758, C759, D3649).

388 Gamma counting is generally carried out using solid detectors since a gas-filled detector will not
 389 provide adequate stopping power for energetic gammas. In solids such as NaI(Tl) or CsI, the
 390 gammas interact by excitation of atoms and energy is transferred to orbital electrons and then
 391 released as light photons when the orbits are refilled. These scintillations are easily detected and
 392 amplified into useable electrical pulses by a multiplier phototube. The NaI(Tl) detector is the
 393 recommended detector for gross gamma counting because of its high efficiency and room
 394 temperature operation.

395 In semiconductor detectors such as Si(Li) and high-purity germanium semiconductors (HPGe),
 396 the gamma photons produce electron-hole pairs and the electrons are collected by an applied
 397 electrical field. A charge-sensitive preamplifier is used to detect the charge transferred and
 398 produce a useable electrical pulse. The semiconductor detectors are widely used in gamma
 399 spectrometry.

400 The output pulses from the multiplier
 401 phototube or preamplifier are directly
 402 proportional to the amount of energy
 403 deposited, which could either be total and
 404 included in the photopeak, or fractional and
 405 included in the continuum or escape peaks,
 406 in the detector by the incident photon. The
 407 pulses may be counted using a scaler or
 408 analyzed by pulse height to produce a
 409 gamma-ray spectrum.

410 Gamma photons interact with the detector
 411 by three distinct processes. The photo-
 412 electric effect results in complete absorption
 413 of the photon energy and produces the full
 414 energy or photopeak shown. The Compton
 415 effect results in a partial absorption of the
 416 photo energy and a scattered photon of
 417 lower energy results. The scattered photon
 418 carries energy away and the Compton continuum results (Figure 15.2). The third interaction is
 419 pair production, which occurs at energies above 1,022 keV and results in the conversion of the
 420 photon to mass as an electron-positron pair. The electron and positron give up their kinetic
 421 energy to the detector and the resulting electron joins the electron population of the detector; the
 422 positron, however, is annihilated in combining with an electron and produces two gamma

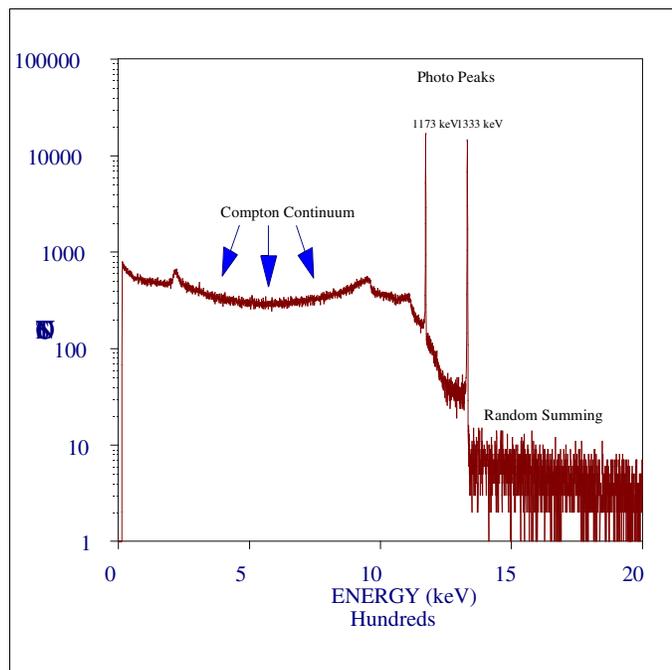


FIGURE 15.2 Gamma-ray Spectra of ⁶⁰Co

423 photons of 511 keV each. One or both of the 511 keV photons may escape from the detector
424 without interacting and the single escape and double escape peaks result.

425 The Comptons, from a higher energy photon, always present an interference problem in the
426 counting of gamma photons and appropriate corrections should be made for this effect. Pair
427 production can also be considered as an interference since the escape peaks may have an energy
428 equal to the lower energy gamma of interest. The Compton and pair production effects can be
429 very significant interferences and should be corrected.

430 The change of the absorption coefficient with gamma energy results in a wide variation of
431 detection efficiency. The detection efficiency falls rapidly as gamma energy increases for a fixed
432 size of detector. Two other important effects are seen as a result of the variation of the absorption
433 coefficient; firstly, low energy photons may be absorbed in massive samples as sample thickness
434 increases, such as large bottles of water, and erroneous results may be obtained. A similar
435 absorption effect is seen in HPGe systems where the can around the detector acts as an absorber
436 for very low-energy gammas and the efficiency passes through a maximum usually around 100
437 keV. The second result is that for low energy gammas a thin detector may be as efficient as a
438 much thicker one since the low-energy gammas are easily stopped in the thin detector.
439 Additionally, thin detectors will have better low energy detection limits because of reduced
440 background interactions.

441 Because of this variation in efficiency and the possible interferences from other activities, gross
442 gamma counting is only reliable when used to compare standards and samples of the same
443 nuclide. The use of gross gamma monitoring systems should be avoided when possible and, in all
444 cases, proper allowance should be made for the lack of accuracy.

445 At high count rates, random sum peaking may occur. Two absorptions may occur within the
446 resolving time of the detector and electronics and are summed and seen as one pulse. For a
447 detector of resolving time, t , and a count rate of A counts per unit time, the time window
448 available for summing is $2At$ (since the count summed could occur as early as t before or as late
449 as t after the other count) and the probability of another count at any time is simply A . Therefore,
450 the sum count rate will be $2A^2t$ in unit time. Random summing is strongly dependent on the
451 count rate A and, if summing occurs, it can be reduced by increasing the sample to detector
452 distance. Modern electronics, both conventional analog and digital (preamplifiers, amplifiers, and
453 analog-to-digital converters) are capable of processing 100,000 cps without any significant lose
454 of resolution. This is because of the very short time constants (resolving time) these systems are
455 capable of producing. Over all detector performance can be affected by count rate because
456 reduced time constants are required which will cause some loss of resolution. When a photon

457 interaction takes place (an event is detected), charge carriers in the form of holes and electrons
 458 are produced. The electrical field produced by the detector's high voltage bias supply causes
 459 these carriers to be swept toward the P and N electrodes of the detector. The time it takes the
 460 carriers to travel to the electrodes is called the "charge collection time." At very high count rates
 461 the detector continues collecting events but the data is not valid. If a second (or third) event takes
 462 place while the first set of charge carriers are still in transit, the energy from the two events get
 463 added together. Therefore, if a 2,000 keV event arrives while a 1,000 keV event is in transit, the
 464 detector would "see" a single 3,000 keV event, producing a random sum peak on pulse pileup.
 465 When the detector starts reporting more sum peaks than valid events, you have exceeded its
 466 count rate capability. Random pulse summing or pileup can also cause peak shape and risetime
 467 problems. But the real upper limit to a detector throughput is pulse summing. This problem can
 468 be reduced or eliminated by either reducing the number of events the detector "sees" by moving
 469 the sample further away, collimate the detector, or use a smaller, less efficient detector; the
 470 smaller the detector the shorter the charge collection time, which means a higher count rate limit.
 471 Peak shifts may also occur with high count rates and short time constants. Another factor that
 472 will affect high count rate performance is improper setting of the amplifier pole zero. Improper
 473 setting of the pole zero with either under or over shooting of input pulse will effect peak
 474 resolution.

475 Well counters that have very high efficiencies are prone to summing since, for a given source
 476 strength, the count rate is higher than for a
 477 detector of lower efficiency. For moderate
 478 and high-source strengths, the trade-off is a
 479 poor one and the well counter is best suited
 480 for low-level work where its high efficiency
 481 is an important advantage.

482 Cascade summing may occur when nuclides
 483 that decay by a gamma cascade are counted.
 484 Cobalt-60 (^{60}Co) is an example; 1,173.2 keV
 485 and 1,332.5 keV from the same decay may
 486 enter the detector and be absorbed, giving a
 487 2,505.7 keV sum peak. Another example of
 488 Cascade summing occurs when counting
 489 sodium-22 (^{22}Na) close to the detector (see
 490 Figure 15.3). Cascade summing may be
 491 reduced and eventually eliminated by
 492 increasing the source-to-detector distance.

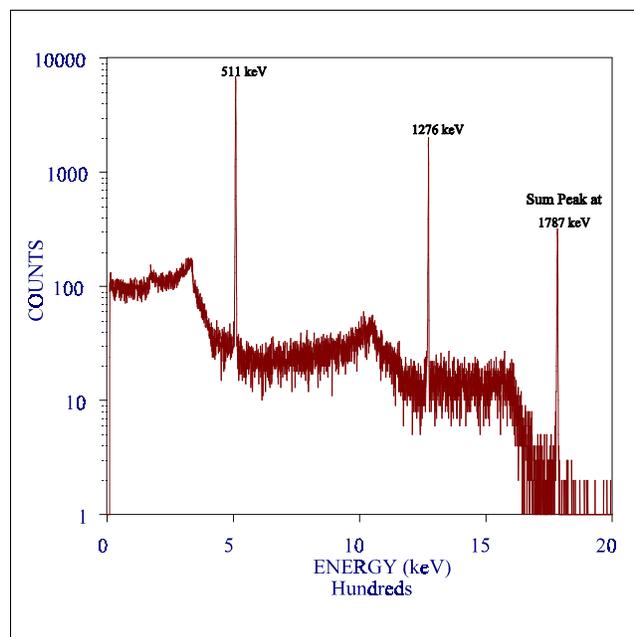


FIGURE 15.3 Energy Spectrum of ^{22}Na

493 The resolution of a gamma detector is the effective limit to its utility even when complex data
 494 reduction methods are used. A typical 76x76 mm NaI(Tl) detector will give full-width half-
 495 maximum (FWHM) of approximately 60 keV at 661.6 keV gamma energy and approximately 90
 496 keV at 1,332.5 keV gamma energy.

497 **15.4.2 Energy Efficiency Relationship**

498 Because of the rapid falloff in gamma
 499 absorption as gamma energy rises, the
 500 detection efficiency shows a similar effect.
 501 Figure 15.4 shows a typical efficiency vs.
 502 energy plot of a 70 percent HPGe p-type, a
 503 35 percent HPGe n-type, and HPGe well
 504 detectors of 122 cm³ with a vespel well and
 505 320 cm³ with a Mg well. The portion of the
 506 curve for n-type and well detectors at low
 507 energies shows that as the absorption
 508 coefficient increases geometry becomes the
 509 limiting factor. The maximum efficiency for
 510 both co-axial detectors is well below 50
 511 percent due to the presence of a beta
 512 absorber, the containment of the detector
 513 and the geometry effect. The p-type detector
 514 shows significant low energy efficiency
 515 drop off because of the absorption of
 516 gamma rays in the detector's inactive Ge dead layer. The well detector shows excellent efficiency
 517 below 100 keV because of the geometry effect and absence of an attenuating germanium dead
 518 layer. The 76x76 mm NaI(Tl) detector is the most widely used size. A large amount of data are
 519 available in the open literature on both the use and results obtained with detectors of this size.
 520 Heath (1964) has written a comprehensive review and supplied many gamma-ray spectra in both
 521 graphical and digital form.

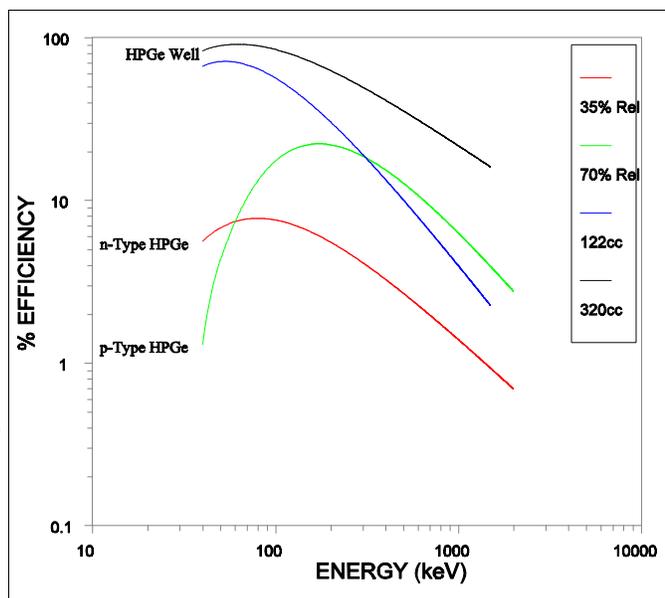


FIGURE 15.4 Efficiency vs. Gamma-ray Energy

522 Other sizes of detectors may be used. However, the following should be noted: smaller detectors,
 523 such as 38x38 mm, will give efficiencies that are low and fall off more rapidly as gamma energy
 524 increases. Small or thin detectors are useful for the measurement of low-energy gammas since
 525 they are less responsive to high-energy gammas and the interference from Compton effects is
 526 reduced. This will result in a lower background.

527 Larger detectors will give higher efficiencies and less falloff as gamma energy increases. Larger
 528 detectors are useful for situations where the highest attainable efficiency is desired and for the
 529 assembly of complete absorption detectors. The increase in efficiency is accompanied by an
 530 increased background count rate and an increase in the probability of summing in the detector.

531 Well detectors will give very high efficiencies, up to about 80 percent for low and moderate
 532 energy gammas. The well detector is useful for low levels of activity and the background of a
 533 well detector is essentially the same as that of a plain cylindrical detector of the same overall
 534 dimensions. Summing becomes a definite problem at high activities since both random and
 535 cascade summing result from the high efficiencies and the high geometry of the well detector.

536 Detector efficiency will also vary as a function of sample geometry. Table 15.1 gives counting
 537 efficiencies obtained with various sample geometries for a 55 percent HPGe detector.

**TABLE 15.1 Typical Percent Gamma-ray Efficiencies for a 55 Percent High-Purity
 Germanium Detector* with Various Counting Geometries**

ENERGY (keV)	FILTER PAPER	50 cm ³ PLANCHET	90 cm ³ AL CAN	600 cm ³ MARINELLI BEAKER
60	15.6	14.6	11.6	5.0
88	15.2	14.2	11.3	7.4
122	15.1	12.6	10.2	8.4
166	12.0	9.6	8.0	7.9
279	9.3	7.4	6.0	6.1
392	7.2	5.5	4.5	4.8
514	5.4	4.2	3.5	3.8
662	4.7	3.6	3.0	3.1
835	3.9	2.9	2.4	2.7
898	3.1	2.4	2.1	2.2
1115	3.0	2.3	1.9	2.1
1173	2.6	2.0	1.7	1.8
1333	2.3	1.8	1.5	1.6
1836	1.7	1.3	1.2	1.3

555 *Although the counting efficiencies listed above were obtained with a 55 percent (relative to a 3x3 inch NaI
 556 detector) HPGe detector, the calculation of counting efficiencies by extrapolation for detectors with different
 557 relative efficiencies is not possible. This is because detectors with the same relative efficiency may be of
 558 significantly different dimensions thus producing a detector/sample solid angle very different than what was used to
 559 prepare this table.

560 **15.4.3 Sodium Iodide Detector Assembly**

561 A cylindrical 76x76 mm NaI detector is activated with about 0.1 percent thallium iodide, with or
562 without an inner sample well, optically coupled to a multiplier phototube, and hermetically
563 sealed in a light-tight container. The NaI(Tl) crystal should contain less than 5 ppm of potassium
564 and be free of other radioactive materials. In order to establish freedom from radioactive
565 materials, the manufacturer should supply a gamma spectrum of the background of the detector
566 between 0.08 and 3,000 keV. The resolution of the detector for the 662 keV gamma from ¹³⁷Cs
567 decay should be less than 50 keV FWHM or less than 7 percent when measured with the source
568 in contact with the end cap.

569 The following components are required for a complete NaI(Tl) gamma-ray spectrometry system:

570 High-Voltage Power Supply 500 to 2,000 V dc regulated to 0.1 percent with a ripple of
not more than 0.01 percent

571 Preamplifier Linear amplifier system to amplify the output from the
multiplier phototube to a maximum output of 10 V.

572 Analyzer with Scaler and Timer A single-channel discrimination system will accept all or
any part of the output from the amplifier and pass it to the
scaler. Any pulses lying outside the preset limits are
rejected. The lower limit is usually referred to as the
threshold and the difference between the two limits is the
window.

Sample mounts and containers may consist of any
reproducible geometry container that is commercially
available. Other considerations are cost, ease of use,
disposal, and effective containment of radioactivity for the
protection of the workplace and personnel from
contamination.

573 Beta Absorber A beta absorber of 3 to 6 mm of aluminum, beryllium, or
poly(methyl methacrylate) should completely cover the
upper face of the detector to prevent betas from reaching the
detector.

574 **15.4.4 High Resolution Germanium Detectors**

575 High resolution germanium detectors are produced from very high purity material, the required
576 level of impurities in the detector crystal is usually less than 10^9 atom/cm³. Any type of
577 germanium—either planar, co-axial or well-configuration—cannot be operated at room
578 temperature because of the large thermally induced leakage current that results. These detectors
579 should be cooled in order to reduce the thermal generation of charge carriers (thus reverse
580 leakage current) to an acceptable level. Otherwise, leakage current induced noise reduces the
581 energy resolution of the detector. The detector is mounted in a vacuum chamber which is
582 attached to or inserted into an liquid nitrogen (LN2) dewar or an electrically powered cooler. The
583 sensitive detector surfaces are thus protected from moisture and condensation contaminants.

584 The boiling point of liquid nitrogen (77 °K) is usually taken advantage of to reduce the operating
585 temperature of the detector. Since germanium detectors can be operated at temperatures as high
586 as 130 °K, mechanical closed-cycle refrigerators can also be used. These systems can cool a
587 detector to as low as 50 °K. Therefore, with proper thermal control the detector can be cooled to
588 its optimum operating temperature. The required preamplifier is normally included as part of the
589 cryostat. In this configuration the preamplifier can also be cooled to reduce electronic noise.

590 HPGe detectors are preferred for the analysis of complex gamma-ray spectra involving many
591 nuclides and peaks. However, for samples with only a few nuclides, the complexity of an HPGe
592 system may not be cost effective. The calibration of germanium detectors is given in ASTM
593 E181.

594 **15.4.5 Low Background High Resolution Germanium Detectors**

595 Environmental samples requiring the lowest possible minimum detection analyses (MDAs)
596 should be counted with large high efficiency germanium detectors in low background cryostats.
597 Most of the background from naturally occurring radionuclides such as ⁴⁰K from building
598 materials, radon decay products, and cosmic rays can be reduced by proper shielding. However,
599 naturally occurring ²³⁵U, ²³⁸U, ²³²Th, and anthropogenic ¹³⁷Cs and ⁶⁰Co may be present in cryostat
600 materials. With careful selection and substitution of materials, low background gamma-ray
601 systems can be fabricated. Germanium crystal mountings and detector end caps have been
602 fabricated with magnesium to eliminate aluminum contaminated with radioactive thorium
603 isotopes. Figures 15.5 and 15.6 show shielded background spectra obtained with 56 percent
604 germanium detectors in standard and extra low background cryostats.

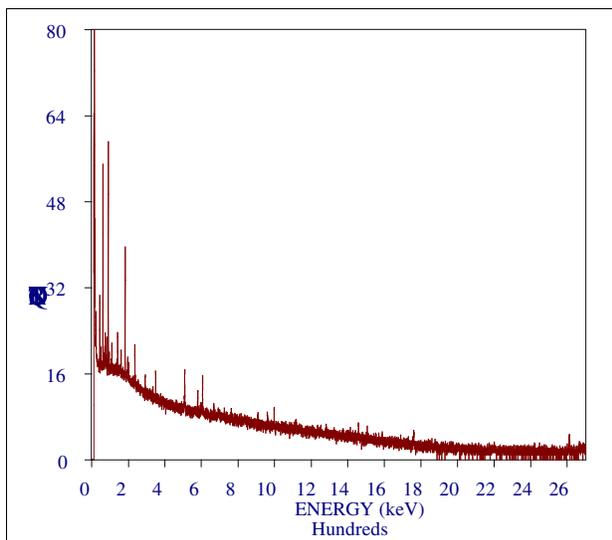


FIGURE 15.5 Standard Cryostat HPGe Background Spectrum

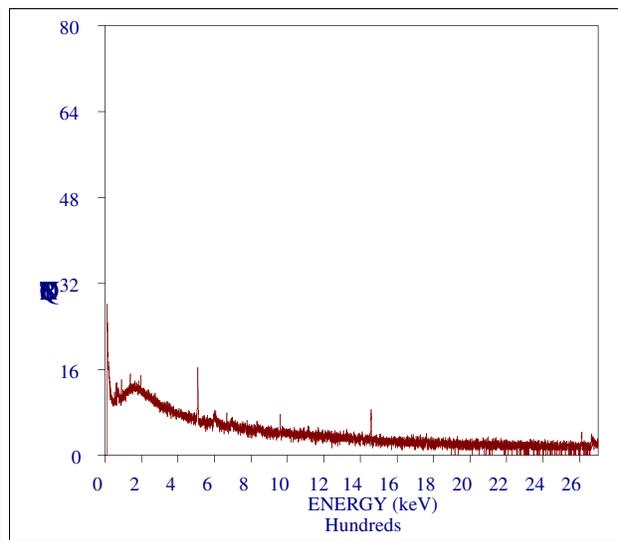


FIGURE 15.6 Low Background Cryostat HPGe Background Spectrum

605 **15.4.6 High Resolution Detectors for Low Energy Spectrometry**

606 High resolution low gamma-ray energy detectors are available in various configurations. The
607 commonly used ones are either high purity germanium or silicon. The various detector types
608 include: planar (Ge or Si), low-energy germanium (LEGe), reverse-electrode germanium (REGe)
609 and extended-range germanium (XtGe). These detectors are equipped with beryllium entrance
610 windows to reduce attenuation. These detectors are especially useful for measuring nuclides that
611 emit gamma or X-rays from a few keV to about 150 keV.

612 **15.4.7 CsI(Tl) Detectors**

613 CsI(Tl) crystals have the highest light output of all known scintillators. However, because light
614 output is not well matched to the sensitivity of the photocathode of a multiplier phototube, the
615 yield for gamma rays is only 45 percent of the efficiency of NaI(Tl). With the proper electronics,
616 CsI(Tl) detectors can be used for α -particle energy discrimination.

617 **15.4.8 CdZnTe Detectors**

618 These gamma-ray detectors, in addition to only being produced in very small volumes, do not
619 have energy resolutions as good as HPGe but are better than NaI(Tl). Their greatest advantage is

620 their ability to operate at room temperature. Because of their small size and resulting low
621 gamma-ray detection efficiency, they are useful for the analysis of very high level sources.

622 **15.4.9 BGO Detectors**

623 Because bismuth germanate ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$) is a high Z, high density (7.13 gcm^3), scintillation
624 material, it is a very efficient gamma-ray absorber. Although BGO crystals have very good
625 peak-to-Compton ratio, their effective efficiency is only 10 to 15 percent as good as a NaI(Tl)
626 crystal. However, BGO is a relatively hard, rugged, non-hygroscopic crystal which does not
627 cleave or absorb any significant amount of the scintillation light. The crystal housing does not
628 require hermetic air-tight sealing. These crystals are useful in applications where high
629 photofraction is required.

630 **15.5 Spectrometry Systems**

631 This section will present a number of different type of detector systems commonly use for
632 gamma-ray spectrometry.

633 **15.5.1 Alpha/Gamma Coincidence Systems**

634 Alpha/Gamma Coincidence Systems have been used for the direct measurement of ^{224}Ra and
635 ^{226}Ra . The counting technique is based upon the coincidence measurement of the characteristic
636 particle-photon emissions of these isotopes. Silver activated zinc sulfide for alpha detection is
637 combined with a NaI well for gamma-ray detection (McCurdy, 1981).

638 **15.5.2 Beta/Gamma Coincidence Systems**

639 Many radionuclides remain in an excited state after what may be considered beta decay. This
640 results in the emission of a gamma ray as the decay process goes to the ground state. A
641 beta/gamma coincidence system will have significantly improved lower limit of detection over a
642 beta or a gamma counting system because of its very low background. Systems have been
643 designed with both $2\text{-}\pi$ and $4\text{-}\pi$ geometry (McCurdy et al., 1980).

644 **15.5.3 Gamma/Gamma Coincidence Systems**

645 These counting systems can provide extremely low backgrounds and are very useful for
646 analyzing those radionuclides that decay with cascading (coincident) gamma rays. The systems
647 usually consist of two large NaI(Tl) detectors with a surrounding active anti-coincidence shield

648 of either NaI(Tl) or plastic phosphor. However, HPGe detectors have also been used in place of
649 the two large NaI(Tl) detectors. Only gamma-ray pulses that are detected in both of the primary
650 detectors at the same time (coincident) and not in the active shield are recorded. Even though
651 these systems can be large, because of the shielding requirements for two detectors and an active
652 annulus, and require complex electronics, the improvement in lower limit of detection for certain
653 radionuclides is worth the investment (Perkins, 1965; Sanderson, 1969).

654 **15.5.4 Photon-Electron Rejecting Alpha Liquid Scintillation Systems**

655 Another technique for the analysis of alpha emitting radionuclides combines liquid scintillation
656 counting with pulse shape discrimination to significantly reduce background counts from photo-
657 electrons produced by ambient background gamma rays and to eliminate interferences from beta
658 emitters in the sample/scintillation cocktail. Pulse shape discrimination electronically selects only
659 pulses produced by alpha particles because of their longer decay times in the scintillation
660 solution. Typical alpha peak resolutions are about 5 percent. Typical detectable activities for
661 alpha emitters such as ^{234}U and ^{241}Am are 0.0037 and 0.37 Bq (0.1 and 10 pCi).

662 **15.6 Special Instruments**

663 This section covers some radiation detection instruments and auxiliary equipment that may be
664 required for special application in the measurement of radioactivity.

665 **15.6.1 4- π Counter**

666 The 4- π counter is a detector designed for the measurement of the absolute disintegration rate of
667 a radioactive source by counting the source under conditions that approach a geometry of 4- π
668 steradian. Its most prevalent use is for the absolute measurement of beta emitters. For this
669 purpose, a gas-flow proportional counter is commonly used. 4- π counting systems consist of two
670 hemispherical or cylindrical chambers whose walls form the cathode, and a looped wire anode in
671 each chamber. The source is mounted on a thin supporting film between the two halves, and the
672 counts recorded in each half are summed.

673 Gamma-ray and hard X-ray counters with geometries approaching 4- π steradian can be
674 constructed from both NaI(Tl) or germanium crystals in either of two ways. A well crystal (that
675 is, a cylindrical crystal with a small axial hole covered with a second crystal) will provide nearly
676 4- π geometry for small sources, as will two solid crystals placed very close together with a small
677 source between them. The counts from both crystals are summed as in the gas-flow counter. The
678 deviation from 4- π geometry can be calculated from the physical dimensions. For absolute

679 gamma-ray counting, the efficiency of the crystal for the gamma energy being measured and the
680 absorption in the detector end cap should be taken into account. The liquid scintillation counter is
681 also essentially a $4\text{-}\pi$ counter for alpha and beta particles, since nearly all the radiations are
682 emitted into and interact with the detecting medium.

683 **15.6.2 Low-Geometry Counters**

684 This type of instrument is particularly useful for the absolute counting of alpha particles. The
685 alpha emitter, in the form of a very thin solid source, is placed at a distance from the detector
686 such that only a small fraction (<1 percent) of the alpha particles are emitted in a direction to
687 enter the counter. This solid angle is obtained from the physical measurements of the instrument.
688 The space between the source and the detector is evacuated to eliminate the loss of alpha
689 particles by absorption in air. The detector can be any counter that is 100 percent efficient for all
690 alpha particles that enter the sensitive volume—a gas-flow proportional counter with a window
691 that is thin (approximately 1 mg/cm^2) compared to the range of the alpha particles or the
692 semiconductor alpha detector with a 1 mg/cm^2 covering. The advantages of this instrument for
693 absolute alpha counting are that the effect of absorption of alpha particles in the source itself is
694 kept to a minimum since only particles that travel the minimum distance in the source enter the
695 detector (particles that have longer paths in the source are emitted at the wrong angle), and back-
696 scattered alpha particles (those that are emitted into the source backing and are reflected back up
697 through the source) lose sufficient energy so that they cannot enter the detector. One such
698 instrument is described in Curtis et al. (1955).

699 **15.6.3 Internal Gas Counters**

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

711 In a modification of the internal gas counter, scintillation counting has been used. The inner walls
712 of the chamber are coated with a scintillation material and the radioactive gas is introduced. An
713 optical window is made a part of the chamber, and the counting is done by placing this window
714 on a multiplier phototube to detect the scintillations. This system is particularly useful for
715 counting radon gas with zinc sulfide as the scintillator. Additional details on internal gas
716 counting may be found in Watt and Ramsden (1964).

717 **15.7 Spectrometers and Energy-Dependent Detectors**

718 The availability of energy-dependent detectors (detectors whose output signal is proportional to
719 the energy of the radiation detected) that are easy to operate and maintain and have good
720 resolution makes it possible to measure not only the total activity of a radioactive sample but the
721 energy spectrum of the nuclear radiations emitted. Nuclear spectrometry is most useful for alpha
722 particles, electromagnetic radiation (gamma and X-rays), and conversion electrons, since these
723 radiations are emitted with discrete energies. Beta spectra have more limited use since beta
724 particles are emitted from a nucleus with a continuous energy distribution up to a characteristic
725 maximum (E- max), making a spectrum containing several different beta emitters difficult to
726 resolve into its components. The advantages of spectrometric over total activity measurements of
727 radioactive sources are increased selectivity, detection limit , and accuracy because nuclide
728 identification is more certain, interference from other radioactive nuclides in the sample is
729 diminished or eliminated, and counter backgrounds are reduced since only a small portion of the
730 total energy region is used for each radiation.

731 The detectors for alpha spectra are gridded ion-chambers and silicon semiconductor detectors.
732 Gridded ion-chambers are no longer available commercially and should be constructed by the
733 user. A variety of semiconductor detectors for alpha spectrometry are commercially available.
734 These detectors have essentially replaced ion-chambers, although the chambers have the
735 advantages of high efficiency (nearly 50 percent) for large-area sources.

736 Silicon alpha particle detectors have a depletion region which is formed by applying a high
737 voltage bias. The electric field produced collects the electron-hole pairs produced by incident
738 alpha particles. Either surface barrier or passivated ion-implanted silicon are commonly used for
739 spectrometry.

740 The principal detectors used for gamma-ray spectrometry are thallium-activated sodium iodide
741 scintillation crystals, NaI(Tl), and high purity germanium semiconductors, HPGe. HPGe
742 detectors are available in n-type and p-type germanium. P-type germanium detectors have dead
743 layers which produce entrance windows from 500 to 1,000 μm thick. On the other hand, n-type

744 detectors have extremely thin entrance windows of about 0.3 μm . These n-type detectors when
745 housed in an end cap with a beryllium window are excellent for measuring both low energy and
746 high energy (3 to 10,000 keV) gamma rays. However, applications which require the best
747 possible energy resolution, peak shape, and efficiency for gamma-ray measurements above 80
748 keV, p-type HPGe is the detector material of choice.

749 For X-rays and very low-energy gamma rays, lithium-drifted silicon semiconductor Si(Li), planar
750 germanium, and gas-filled thin window (approximately 1 mg/cm²) proportional counters are
751 used.

752 The electronic version of Heath's (1964) Ge(Li) and Si(Li) Detector Gamma-ray Spectrum
753 Catalogue is available in two forms. The document is on the Web at <http://id.inel.gov/gamma>; it
754 is also available on a CD-ROM.

755 The portion of the crystal end cap through which gamma rays enter is normally thinner, or
756 constructed of a low-Z material, like beryllium or magnesium, than the rest of the package in
757 order to reduce low-energy attenuation. Sodium iodide crystals are available in a large range of
758 sizes and shapes, from thin crystals for X-ray analysis and small 25 by 25 mm cylinders to
759 hemispheres and cylinders over 300 mm in diameter. Information on the types of crystal
760 packages and mountings is available from the manufacturers.

761 A complete NaI(Tl) detector spectrometer requires a high-voltage power supply for the phototube
762 (usually operated at 600 to 1,000 V), a preamplifier, linear amplifier, pulse-height analyzer, and
763 output recorder. Because NaI(Tl) detectors cannot resolve gamma-ray energies that are only a
764 few keV apart, a least-squares computer program should be used to quantify a complex gamma-
765 ray spectrum.

766 Germanium and silicon detectors are junction-type semiconductor devices. With silicon
767 detectors, a sensitive region is produced by drifting lithium under the influence of an electric
768 field at an elevated temperature (100 to 400 °C) into the crystal. The crystal then functions as a
769 solid ion chamber when a high voltage is applied. Today, germanium detectors are made with
770 very high purity material that does not require lithium drifting. In order to obtain high resolution,
771 these detectors should be operated at low temperatures to reduce thermal noise. At room
772 temperature, sufficient free electrons will be present in the crystal to obscure the measurement of
773 gamma and X-rays. Consequently, the detectors are operated at liquid nitrogen temperatures by a
774 cryostat consisting of a metallic cold-finger immersed in a Dewar flask containing liquid nitrogen
775 or mechanically refrigerated.

776 The electronic components required to obtain spectra are similar to those for NaI(Tl) detectors,
777 except that because smaller pulses should be measured, high-quality electronics should be used.
778 A complete HPGe system includes a high-voltage bias supply for the detector, a preamplifier,
779 amplifier (usually charge-sensitive), pulse height analyzer, and recording device. With the
780 exception of extremely complex spectra, most high resolution spectra can be quantified by
781 simple integration of full energy gamma-ray peaks.

782 The resolution of gamma-ray detectors
783 is usually specified in terms of its
784 FWHM. Detector resolution, expressed
785 in percent, improves with increasing
786 energy and for NaI(Tl) detectors and is
787 usually determined from the 662 keV
788 gamma ray emitted in the decay of ^{137}Cs .
789 This is shown graphically in the
790 gamma-ray spectrum in Figure 15.7. For
791 HPGe detectors, ^{60}Co is measured
792 25 cm above the detector end cap.
793 Quality sodium iodide crystals have
794 resolutions in the range of 6.5 to 7
795 percent for ^{137}Cs . Detection efficiency
796 for the same geometry and window
797 thickness is a function of several
798 parameters and much published
799 information on efficiencies for various
800 energies, detector sizes, source-to-detector distances, and other variables is available
801 (Crouthamel et al., 1970). The efficiency for gamma-ray detection is expressed as full energy
802 peak efficiency; the fraction of incident gamma rays that give a full-energy peak for a particular
803 source-detector configuration. For a 102 mm thick NaI(Tl) crystal, with the source on the surface
804 (zero distance), this fraction is approximately 0.24 for the 661.6 keV gamma ray of ^{137}Cs and
805 approximately 0.14 for the 1,332.5 keV gamma ray of ^{60}Co . The peak-to-valley or peak-to-
806 Compton ratio is the ratio of counts at the maximum height of the full-energy peak to the counts
807 at the minimum of the Compton continuum. A high ratio indicates narrow peaks, that is, good
808 resolution, for that particular energy.

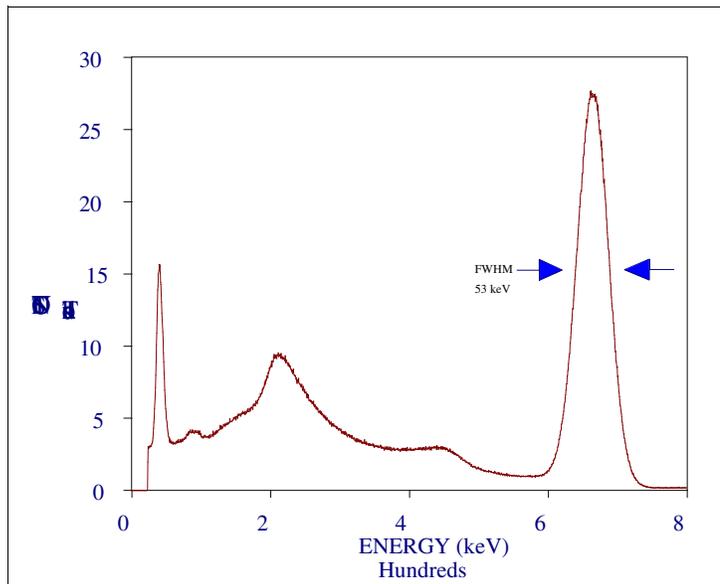


FIGURE 15.7 NaI(Tl) Energy Spectrum of ^{137}Cs

809 The efficiency specification of a HPGe detector is expressed by comparing its ^{60}Co , 1,332.5 keV
810 efficiency at 25 cm with that of a 76x76 mm cylindrical NaI(Tl) detector at the same distance.

811 Photopeaks are spread over a much
 812 smaller energy range in germanium
 813 than in sodium iodide, the background
 814 under the peak is much less (Figure
 815 15.8). This means that for small
 816 sources of moderately energetic
 817 gamma rays, germanium is more
 818 sensitive than sodium iodide.

819 Typical specifications for a
 820 germanium gamma-ray detector could
 821 include but should not be limited to
 822 the following:

823 DETECTOR: The gamma-ray
 824 detector should consist of High-
 825 Purity n-type germanium.

826 SIZE: The germanium crystal
 827 should be at least 5.5 cm in diameter and at least 7.0 cm long.

828 EFFICIENCY: The relative counting efficiency compared to a 3"x3" NaI detector at 25 cm for
 829 ⁶⁰Co (1,332 keV) should be equal to or better than 50 percent.

830 RESOLUTION: The resolution (FWHM) of the detector should be equal to or better than
 831 2.2 keV at 1,333 keV (⁶⁰Co). The resolution (FWHM) at 122 keV (⁵⁷Co) of the detector
 832 should be equal to or better than 1.0 keV. The detector resolution at FWTM should be equal
 833 to or better than 2 times the FWHM.

834 PEAK-TO-COMPTON RATIO: The peak-to-Compton ratio for 1,333 keV (⁶⁰Co) should be equal
 835 to or better than 50:1.

836 BACKGROUND: Low radioactivity materials should be used so that any full energy gamma-ray
 837 line (excluding 511 keV and 1,460 keV) present in a 1,000-minute background spectrum
 838 (100-2,000 keV) obtained in a graded 10 cm lead shield should not exceed 0.20 counts per
 839 minutes.

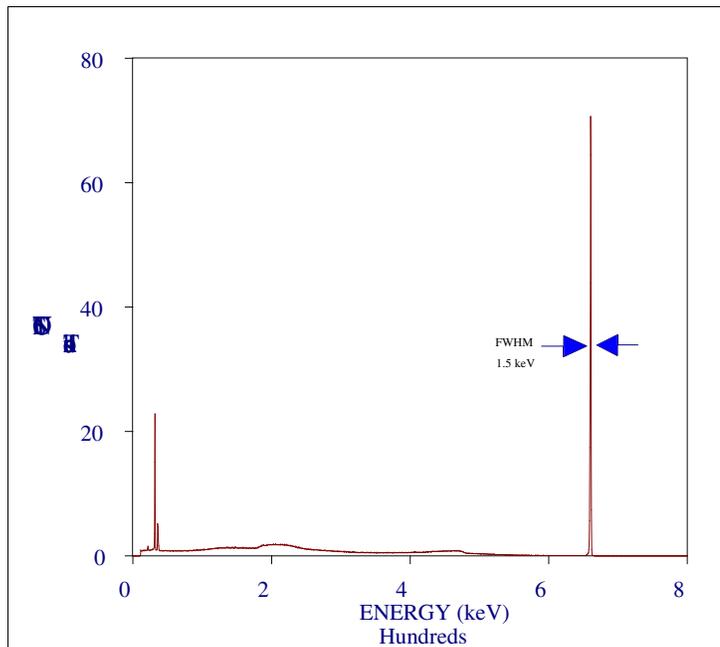


FIGURE 15.8 HPGe Energy Spectrum of ¹³⁷Cs

840 CONTACTS: The internal detector contacts should be DC-coupled ion implants so that low
841 energy gamma-ray attenuation is avoided.

842 PREAMP: A low-noise, cooled field-effect transistor preamplifier should be used to provide
843 the detector output signals.

844 CRYOSTAT: The cryostat should be constructed of low radioactivity materials throughout and
845 should contain sufficient lead shielding in order to minimize radiation from the dewar or
846 lower portion of the cryostat.

847 END CAP: The end cap should consist of a 20 mil beryllium window with 0.5 mm aluminum
848 side walls and be no greater than 7.6 cm diameter (OD). This diameter should be maintained
849 for at least 8 cm from the end cap. Below this point the outside diameter of the end cap may
850 be increased. The top of the end cap should be between 95 and 102 cm above the outside base
851 of the dewar.

852 TEMPERATURE: The cryostat should contain a temperature sensing circuit to provide high
853 voltage shut down in order to prevent preamplifier damage in case of warm-up due to loss of
854 liquid nitrogen.

855 Spectra of beta particles and conversion electrons can be obtained with sodium iodide and n-type
856 HPGe detectors. A germanium detector with a volume of 120 cm³ has an efficiency approxi-
857 mately 20 percent that of a 76x76 mm NaI(Tl) crystal. Larger HPGe detectors are available with
858 relative efficiencies over 150 percent when compared with a 76x76 mm NaI(Tl) crystal.
859

860 Presently available germanium detectors have resolutions of 1.5 to 2.5 keV at 1,332.5 keV. The
861 method used to measure the energy resolution is described in ANSI/IEEE 325. This greater
862 resolution makes this detector the one of choice for gamma-ray spectrometry and cancels to some
863 extent the higher efficiency available from sodium iodide. Since the pulses from a single
864 semiconductor detectors sufficiently thick (a few centimeters) to absorb the particles completely.
865 One disadvantage of sodium iodide detectors is their relatively thick entrance windows. Other
866 semiconductor detectors have thin beryllium entrance windows and can be used for beta
867 spectrometry.

868 Good spectra of low-energy beta particles, conversion electrons, and X-rays can be obtained with
869 a gas-flow proportional Counter provided that a linear preamplifier is used. The resolution is
870 intermediate between NaI(Tl) and HPGe. Organic scintillators, such as anthracene and
871 polystyrene polymerized with scintillating compounds, are also useful for beta spectrometry.

872 They are packaged with a phototube in a
 873 manner similar to sodium iodide
 874 crystals. Liquid scintillation mixtures
 875 also give beta spectra, and the output of
 876 a commercial liquid scintillation counter
 877 is usually fed into a multichannel pulse-
 878 height analyzer to obtain a beta energy
 879 spectrum (Blanchard et al., 1960). A
 880 spectrum of ^{210}Pb , ^{210}Bi , and ^{210}Po in
 881 Figure 15.9 shows the resolution
 882 obtainable by liquid scintillation
 883 counting of aqueous samples in a
 884 dioxane-based solution. The ^{210}Bi curve
 885 is from a beta particle, and the ^{210}Po
 886 peak is from an alpha particle. Organic
 887 scintillators are preferable to sodium
 888 iodide for beta spectrometry because
 889 less back scattering occurs.

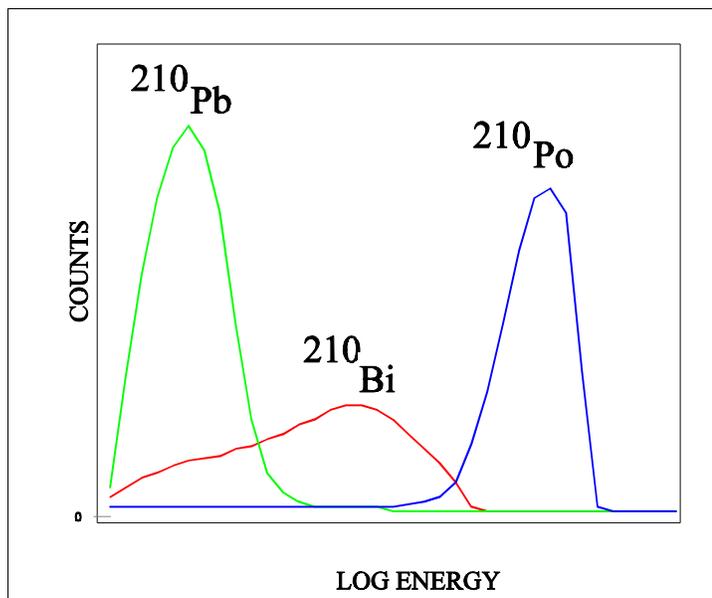


FIGURE 15.9 Spectrum of ^{210}Pb , ^{210}Bi , and ^{210}Po

890 15.7.1 Anti-Coincidence Counters

891 Substantial background reduction can be achieved in beta and gamma counters by surrounding or
 892 covering the sample detector with another detector also sensitive to beta or gamma radiation, and
 893 connecting them electronically so that any pulse appearing in both detectors at the same time is
 894 canceled and not recorded as a count. This is referred to as anti-coincidence shielding, and is
 895 recommended for obtaining very low backgrounds. This type of counter was used for many years
 896 in directional studies of cosmic rays, and was first applied to reducing the background of beta
 897 counters by Libby in his study of natural ^{14}C . The thick metal shielding (lead, iron, or mercury)
 898 ordinarily used to reduce cosmic-ray and gamma-ray background should also be present, and is
 899 placed outside the anti-coincidence shielding.

900 Anti-coincidence shielding of gamma-ray detectors operates in a similar way, and is particularly
 901 useful in reducing the Compton continuum background of gamma rays (Nielson, 1972). Gamma
 902 rays that undergo Compton scattering and produce a pulse in both the detector and the anti-
 903 coincidence shield are canceled electronically. Ideally, only those gamma rays that are completely
 904 absorbed in the sample detector produce a count that is recorded with the total energy of the
 905 gamma ray (full-energy peak). There are second-order effects that prevent complete elimination

906 of Compton scattering, but the improvement is substantial (Perkins, 1965, and Cooper et al.,
907 1968).

908 **15.7.2 Coincidence Counters**

909 In coincidence counting, two or more radiation detectors are used together to measure the same
910 sample, and only those nuclear events or counts that occur simultaneously in all detectors are
911 recorded. The coincidence counting technique finds considerable application in studying
912 radioactive decay schemes; but in the measurement of radioactivity, the principal uses are for the
913 standardization of radioactive sources and for counter background reduction.

914 Coincidence counting is a very powerful method for absolute disintegration rate measurement
915 (Friedlander et al., 1964; IAEA, 1959). Both alpha and beta emitters can be standardized if their
916 decay schemes are such that β - γ , γ - γ , β - β , α - β , or α -X-ray coincidence occur in their decay.
917 Gamma-gamma coincidence counting with the source placed between two sodium iodide
918 crystals, is an excellent method of reducing the background from Compton scattered events. Its
919 use is limited, of course, to counting nuclides that emit two photons in cascade (which are
920 essentially simultaneous), either directly as in ^{60}Co , by annihilation of positrons as in ^{65}Zn , or by
921 immediate emission of a gamma ray following electron capture decay. Non-coincident pulses of
922 any energy in either one of the crystals will be canceled, including cosmic-ray photons in the
923 background and degraded or Compton scattered photons from higher energy gamma rays in the
924 sample. Thus, the method reduces interference from other gamma emitters in the sample. When
925 two multichannel analyzers are used to record the complete spectrum from each crystal, singly
926 and in coincidence, then the complete coincident gamma-ray spectrum can be obtained with one
927 measurement. The efficiency for coincidence counting is low since it is the product of the
928 individual efficiencies in each crystal, but the detection limit is generally improved because of
929 the large background reduction (Nielsen and Kornberg, 1965). This technique is often referred to
930 as two-parameter or multidimensional gamma-ray spectrometry.

931 Additional background improvement is obtained if the two crystals are surrounded by a large
932 annular sodium iodide or plastic scintillation crystal connected in anti-coincidence with the two
933 inner crystals. In this case a gamma ray that gives a pulse, but is not completely absorbed in one
934 of the two inner crystals, and also gives a pulse in the surrounding crystal, is canceled
935 electronically (Perkins, 1965, and Nielsen and Kornberg, 1965). This provides additional
936 reduction in the Compton scattering background. Germanium detectors may be used in place of
937 the inner sodium iodide crystals for improved resolution and sensitivities (Cooper et al., 1968).
938 An example of an assay for plutonium content using passive thermal-neutron coincidence

939 counting is given in ASTM (C1207). Another example of passive thermal-neutron coincidence
940 counting using a moveable californium source is given in ASTM (C1316).

941 **15.8 Shielding**

942 The purpose of shielding is to reduce the background count rate of a measurement system.
943 Shielding reduces background by absorbing some of the components of cosmic radiation and
944 some of the radiations emitted from material in the surroundings. Ideally, the material used for
945 shielding should itself be free of any radioactive material that might contribute to the
946 background. In practice, this is difficult to achieve as most construction materials contain at least
947 some naturally radioactive species (such as ^{40}K , members of the uranium and thorium series,
948 etc.). The thickness of the shielding material should be such that it will absorb most of the soft
949 components of cosmic radiation. This will reduce cosmic-ray background by approximately 25
950 percent. Cosmic-ray interactions in lead shields will produce lead X-rays that are in turn shielded
951 by cadmium and copper liners. Such a shield is referred to as a “graded shield.” Six millimeters
952 of oxygen-free high-conductivity (OFHC) copper can also be used to reduce the cosmic-ray
953 produced lead X-rays without the cadmium liner. Shielding of beta- or gamma-ray detectors with
954 anti-coincidence systems can further reduce the cosmic-ray or Compton scattering background
955 for very low-level counting.

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

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

963 The size and interior dimensions of shields constructed for gamma-ray spectrometry or gamma
964 counting in general should be considered so that sample back scatter radiation from the shield
965 wall to the detector is minimized. In general, shield wall should be at least 10 cm from the
966 detector. Back scatter radiation will fall off as the square of the detector to shield wall distance.

967 **15.9 Instrument Calibration**

968 Calibrations of instruments should be made using reference materials of known and documented
969 value and stated uncertainty. These reference materials should be supplied by:

- 970 • National Institute of Science and Technology (NIST) directly;
- 971 • A standard source supplier whose measurement capabilities and/or manufacturing processes
972 are periodically tested by NIST; and
- 973 • A user who documents derived materials with stated uncertainty and whose value has been
974 verified with analytical and measurement systems that have been periodically tested through
975 an unbroken chain of comparisons to the national physical standards.

976 Periodic testing of source manufacturers, whether they be commercial or agency suppliers or end
977 users, is most cost effectively implemented through measurement assurance programs that are
978 ultimately linked to NIST traceability (Hoppe, 1990).

979 A comprehensive discussion of germanium detector set up and calibration can be found in ANSI
980 N42.14.

981 **15.10 Other Considerations**

982 **15.10.1 Alpha**

983 15.10.1.1 Troubleshooting

984 A number of factors can influence alpha counting results. These include attenuation or self
985 absorption, detector contamination, and other radionuclide interference. Attenuation or self
986 absorption corrections need not be made if constant conditions are maintained for sample and
987 calibration standard counting. If conditions can not be held constant, then corrections will have to
988 be made in order to produce accurate results. For example, the gamma rays from ^{137}Cs in a water
989 matrix counted in a 90 cm³ aluminum can will require a 15 percent correction. Individual
990 electrical line conditioners or uninterruptible power supplies as well as supplemental air
991 conditioning can be provided in the counting rooms to maintain electrical and environmental
992 stability. Additionally, humidity control can also provided. Temperature and humidity may be
993 recorded with a chart recorder.

994 Detector contamination can also be a problem in some cases and, therefore, detector backgrounds
995 should be periodically checked. Contaminated detectors will have higher background counts and
996 even when sample spectra are corrected for the presence of contamination the higher background
997 results in higher MDAs. Finally, some alpha counters may be sensitive to beta radiation, and
998 corrections may have to be made for this interference. For a routine operating alpha counting

999 system periodic instrument QC checks should be performed at some specified frequency. This
1000 would include, as appropriate, counting efficiency, background, resolution, gain, and voltage
1001 plateau.

1002 Solid state detectors used for alpha spectrometry can become contaminated by recoil. This recoil
1003 contamination, which increases the detector background, takes place when fragments from
1004 sources travel to the detector and are implanted in the detector surface by the recoil energy
1005 imparted to the nucleus of an alpha-emitting atom. The energy of the fragments may be sufficient
1006 to implant them in the detector so that they cannot be removed non-destructively. Recoil
1007 contamination can invalidate a count after only a single sample count and cause a constant need
1008 to decontaminate equipment.

1009 The application of a negative bias to the sample, in conjunction with an absorbing layer of air, or
1010 a thin film absorber ($12 \mu\text{g}/\text{cm}^3$) helps to keep recoil particles from imbedding themselves into
1011 the detector. For better resolution and where recoil contamination is of no concern, it is advisable
1012 to maintain a low pressure. Typically, systems can pump down to under $50 \mu\text{m}$ and, by
1013 continuously running the pump, maintain that level indefinitely.

1014 Detector contamination dominated by two processes, alpha recoil and “volatilization” of
1015 polonium. Alpha recoil contamination occurs when an alpha-emitting nuclide on the source plate
1016 decays to an alpha-emitting daughter or string of progeny. Since the specific activity is inversely
1017 proportional to the half-life for a fixed number of atoms, recoil will produce the most background
1018 activity when relatively short-lived progeny are produced. However, if the half-lives in question
1019 are very short (say up to a few hours), they will decay away quickly enough to be of little concern
1020 in alpha spectrometry. Particularly serious are those cases that involve transfer of recoil progeny
1021 with half-lives from days to weeks, short enough that a reasonable amount of parent activity will
1022 produce a significant amount of recoil contamination, and long enough that decay back to normal
1023 background levels will require an inappropriately long time. In addition, the effect is chronic:
1024 similar recoil-producing samples counted in the same chamber will produce a long-term build-up
1025 of detector background which could eventually become serious.

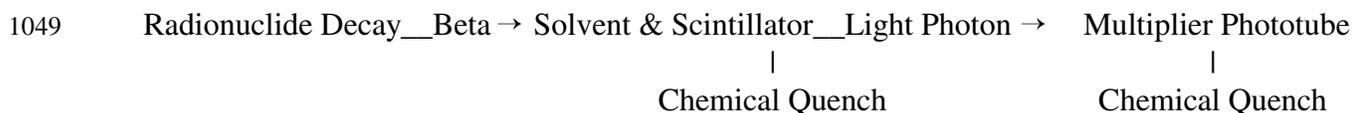
1026 Some common examples of decay-chains that produce recoil contamination include ^{228}Th , ^{229}Th ,
1027 and ^{226}Ra . It is important to realize that even β -emitting nuclides ejected by alpha recoil can
1028 contribute to alpha background if they subsequently decay to alpha emitters. For example, the
1029 direct daughter of ^{229}Th is ^{225}Ra which decays by β -emission to the α -producing daughter ^{225}Ac .

1030 Contamination of detectors by polonium isotopes, such as ^{210}Po ($t_{1/2} = 138.4$ days), should occur
1031 by some other process than alpha recoil. Note that ^{210}Po , the last radioactive member of the ^{238}U

1032 decay series, is the daughter of ^{210}Bi , a beta-emitter. The transfer of polonium from a source to a
1033 silicon detector has been attributed to “aggregate” recoil and inherent volatilization of polonium
1034 at low pressure. Whatever the actual cause, it is clear that polonium activity is indeed transferred
1035 to detectors, a very serious problem with long-lived ^{210}Po and even worse when working with
1036 ^{209}Po ($t_{1/2} = 102$ years) as a yield tracer.

1037 Liquid Scintillation Quenching

1038 Quenching, which is probably the most prevalent interference in liquid scintillation counting, can
1039 be defined as anything which interferes with the conversion of radionuclide decay energy to
1040 photons emitted from the sample vial, resulting in a reduction of counting efficiency. Two types
1041 of quenching may be encountered in liquid scintillation counting: optical or color quenching and
1042 chemical quenching. Color quenching results in a reduction of the scintillation intensity (as seen
1043 by the multiplier phototubes) due to absorption of the scintillation light by materials present in
1044 the scintillation solution resulting in fewer photons per quanta of particle energy and a reduction
1045 in counting efficiency. Chemical quenching results in a reduction in the scintillation intensity due
1046 to the presence of materials in the scintillation solution that interfere with the process leading to
1047 the production of light resulting in fewer photons per quanta of particle energy and a reduction in
1048 counting efficiency. The quenching process may be illustrated as follows.



1050 One can have both types of quenching present in a sample. Note that in chemical quenching all
1051 energy radiations are equally effected, but in color quenching not all energy radiations are equally
1052 effected. Therefore, the measured sample counts should be corrected for quenching effects so
1053 that the radioactivity in the sample can be quantified. Typical quench corrections include
1054 Channels Ratio, External Standard and Internal Standardization.

1055 Attenuation

1056 Attenuation or self absorption corrections may be necessary for alpha counting. Attenuation
1057 corrections should be made whenever the sample matrix differs from that of the calibration
1058 standard. For example, when a gross alpha analysis is performed on an evaporated water sample
1059 of some thickness and an electroplated standard was used for the calibration. Attenuation
1060 corrections will have to be made. Alpha particle attenuation corrections will generally be
1061 necessary with a sample density thickness greater than about one mg/cm^2 .

1062 Figure 15.10 shows how severe the attenuation of alpha particles is in air.

1063 15.10.1.2 Calibration

1064 Alpha counting instrumentation should
 1065 be calibrated with the specific radionuclide of interest or a radionuclide of
 1066 similar alpha energy under the same
 1067 configuration that the sample will be
 1068 counted. The standard should contain
 1069 the same solid material as the sample
 1070 and be of the same weight. If the
 1071 samples and calibration standard are not
 1072 counted under identical conditions, then
 1073 corrections will have to be made. Also,
 1074 if there is a variation in weight from
 1075 sample to sample corrections will have
 1076 to be made, typically a calibration curve
 1077 relating sample weight to counting
 1078 efficiency is used.
 1079

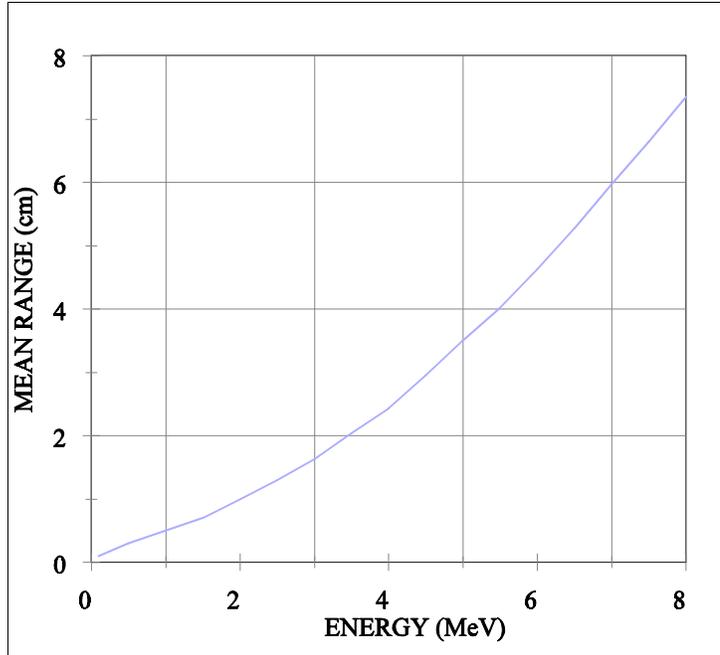


FIGURE 15.10 Range vs. Energy for Alpha Particles in Air

1080 Alpha calibration standards are available from NIST or NIST-traceable commercial vendors.
 1081 Among the radionuclides available are ^{230}Th , ^{241}Am , ^{235}U , ^{239}Pu , ^{228}Th , ^{238}U , and ^{226}Ra . Other
 1082 radionuclides are also available, NIST or a commercial vendor should be contacted regarding
 1083 procurement. Sources should be prepared in the manner in which the sample will be counted.
 1084 The source may be procured as a solution and then prepared in the appropriate counting
 1085 geometry, or the source may be procured directly in the appropriate geometry, such as an
 1086 electroplated standard.

1087 15.10.1.3 Costs

1088 There are three major types of detectors used for alpha counting. Their cost will depend on the
 1089 type of information wanted and the number of detectors in the unit.

1090 Solid state silicon surface barrier detectors are used to count and distinguish alpha particles of
 1091 different energies. An alpha spectrometer consists of a vacuum chamber, detector, electronics to
 1092 amplify the signal, a multichannel analyzer, and some means of collecting data. A system with

Nuclear Counting Instrumentation

1093 eight detectors, vacuum pump, computer and the software necessary for data collection and data
1094 reduction costs approximately \$50,000.

1095 A liquid scintillation counter can be used to count alpha particles and in some cases provide
1096 some information about the energy distribution, although with poorer resolution than silicon
1097 surface barrier detectors. The LSC unit is typically set up to count samples sequentially, using
1098 one detector and automatic sample changer. The price depends on the background required, and
1099 will range from \$25,000 to as much as \$45,000. This price includes a computer and the
1100 appropriate software.

1101 A gas-flow proportional counter is used to count samples for a gross alpha (or beta) activity. The
1102 price of a unit depends on the number of detectors, the size of each detector, and the accessories.
1103 One major accessory could be an automatic sample changer. A system with 8 to 10 small
1104 detectors (1 inch in diameter) will cost from \$35,000 to more than \$60,000.

1105 There are no maintenance costs associated with an alpha spectrometer. If properly used and
1106 monitored, the system will retain its specifications for a long time. The detectors may need
1107 replacing eventually, if its resolution deteriorates or it becomes contaminated, at a cost of \$500-
1108 \$1,000 each.

1109 A liquid scintillation counter requires the use of an organic scintillation cocktail, which cannot be
1110 reused. The total cost of this cocktail, combined with the cost of the sample vials, should not
1111 exceed \$500 for an annual throughput of approximately 1,000 samples.

1112 The operation expense associated with the use of a gas-flow proportional counter is for the ultra-
1113 high purity P10 gas, which is necessary if stable efficiencies and low backgrounds are required.
1114 All proportional counters should have calibrated gas regulators for accurate and reproducible
1115 settings of flow rates. The flow rate should be placed with the QC information that is with the
1116 other instrument QC. For almost constant operation of a system with eight detectors, as many as
1117 24 tanks of P10 gas per year will be required, at a total cost of approximately \$7,000.

1118 All of the above instruments should be in a fairly constant temperature and low humidity
1119 environment, so that air conditioning and/or heating costs need to be factored in, as needed.

1120 15.10.1.4 Quality Control

1121 Statistical quality control (SQC) is discussed here to familiarize the reader with its application to
1122 nuclear counting instrumentation. More detailed information about SQC is provided in
1123 Chapter 19.

1124 The primary tool for statistical quality control is the control chart. A control chart is a graphical
1125 tool for monitoring the distribution of values produced by a measurement process or system. The
1126 distribution of values observed during a period when the system is in statistical control is used to
1127 set up the control chart. Subsequent values are then plotted on the chart and inspected to ensure
1128 that the system remains in control.

1129 Typically one or more control charts for counting efficiency and background are maintained for
1130 each counting instrument. The instrument should be fully operational before the control charts
1131 are implemented. However, control charts should be in use before calibration of the instrument
1132 for a particular analysis to ensure that the instrument parameters are in statistical control during
1133 the calibration.

1134 The selection of the check source for monitoring counting efficiency is critical and should be
1135 made after considering guidance in this document. The source geometry, half-life, and radiation
1136 energy are important factors.

1137 A control chart should be based on an initial data set obtained from at least 15 measurements.
1138 Ideally, at least 10,000 counts per measurement are recommended to provide a relative counting
1139 uncertainty of no more than 1 percent. For some instruments, achieving the recommended 10,000
1140 counts may be impractical, especially for a background control chart. It may also be undesirable
1141 to place a high-activity efficiency check source in a low-background detector because of the
1142 potential for contamination.

1143 The initial measurements should represent the measurement system as it is used over time.
1144 Making the measurements over several days ensures that variability due to temperature and
1145 humidity changes is included. The source should be repositioned before each measurement to
1146 ensure that variability due to positioning error is included.

1147 The mean and standard deviation of the counts or count rates are estimated from the initial data
1148 set. The mean is used as the central line (CL) of the control chart. Warning limits are placed at ± 2
1149 standard deviations from the central line, and control limits are placed at ± 3 standard deviations
1150 from the central line.

1151 Statistical tests of the data distribution should be performed at the time the warning and control
1152 limits are established. Tests for normality are common. It is also common to test whether the
1153 counts follow the Poisson model (Chapter 19).

1154 The central line and warning and control limits for an efficiency control chart should be adjusted
1155 for radioactive decay of the check source unless the source is very long-lived. Either the limits or
1156 the data points may be decay-corrected. It may also be necessary to adjust the counting time for
1157 the check source measurements if the source decays considerably during the period when the
1158 chart is in use. It is important to note that the relative standard deviation of the measured values
1159 increases as the mean number of counts per measurement decreases.

1160 When a measured value falls within the warning limits, the measurement system is considered to
1161 be in control. If a value falls outside the control limits, the system is considered out of control.
1162 These two rules are commonly used to evaluate control charts, although stricter evaluation
1163 criteria are sometimes used. Common sense should be exercised if the data meet the objective
1164 evaluation criteria but nevertheless demonstrate patterns or trends that might indicate developing
1165 problems. For example if a long increasing or decreasing sequence of values is observed, an
1166 investigation is probably warranted even if all of the values are between the warning limits.

1167 Generally, if a value falls within the control limits but outside the warning limits, the system may
1168 require more attention but it is not yet considered definitely out of control. The Westgard Rules,
1169 which are recommended by ASTM E1329, provide more elaborate criteria for evaluating such
1170 measurements.

1171

The Westgard Rules*
1. Is the measurement more than 2 sigma from the mean? If not, go to Step 7.
2. Is the measurement more than 3 sigma from the mean? If so, go to Step 8.
3. Are the last two measurements more than 2 sigma from the mean? If so, go to Step 8.
4. Is the range of the last two measurements more than 4 sigma? If so, go to Step 8.
5. Are the last four measurements more than 1 sigma from the mean? If so, go to Step 8.
6. Are the last ten measurements more on the same side of the mean? If so, go to Step 8. Otherwise, go to Step 7.
7. Accept the measurements. Stop.
8. The measurements are out of control. Stop.

1180 *Adapted from ASTM E1329.

1181 The following two sections on proportional counting and liquid scintillation counting are
1182 applicable to both alpha and beta measurements.

1183 *Proportional Counters*

1184 The following should be considered when QC checks are not within limits.

- 1185 1. Is the standard decay corrected, correctly?
- 1186 2. Check log book to see what changes were made to counter and if the repairman recently
1187 changed any switch settings.
- 1188 3. If gas cylinder was changed recently, was system allowed to purge? Was correct gas (¹⁰P)
1189 obtained? Verify the correct regulator pressure, and ensure the gas cylinder valve is open all
1190 the way.
- 1191 4. If backgrounds are high, check for dirt or dust on the background planchet. Check window
1192 for contamination and replace if necessary.
- 1193 5. Check alpha and beta voltages.
- 1194 6. Check discriminator settings.
- 1195 7. Check voltages on nim bin power supply ($\pm 12V$, $\pm 24V$).
- 1196 8. Check alpha and beta plateau voltage for drift.

1197 *Liquid Scintillation Counters*

1198 The following should be considered when QC checks are not within limits.

- 1199 1. Is the standard decay corrected, correctly?
- 1200 2. Has the quench value for the unquenched standard for the instrument changed? The quench
1201 value for the unquenched standard indicates the overall gain of the system. Run the
1202 autocalibration and verify the result with the historical result.
- 1203 3. Check for dirt or fingerprints on outside of vial.
- 1204 4. Check for dirt inside instrument.

- 1205 5. Is sample two phase?
- 1206 6. Has standard dark adapted and reached temperature equilibrium?
- 1207 7. Check log book to see what changes were made to machine and if repairman recently
1208 changed any switch settings.

1209 **15.10.2 Beta**

1210 15.10.2.1 Introduction

1211 Accurate beta particle measurements will depend upon the degree and extent to which the
1212 parameters that affect the measurement process under considerations are quantified. These
1213 parameters may include:

- 1214 • Radiation detector used;
- 1215 • Material and shape of the final sample mount;
- 1216 • Form and thickness of final sample for analysis;
- 1217 • Radionuclide purity of final sample;
- 1218 • Final sample-to-detector distance; and
- 1219 • Beta particle energy.

1220 Beta particle attenuation or self absorption corrections to the detector efficiency may be
1221 necessary depending on the beta particle energy detection system and final sample form. The
1222 potential of detector contamination from sample measurements is a function of the type of
1223 detector used and the stability of the final sample composition. The inherent beta particle
1224 background of the various detection systems should be evaluated and its contribution removed
1225 from the sample measurement result. The beta particle measurement system should be calibrated
1226 with NIST-traceable standards and its subsequent performance held to established measurement
1227 quality requirements through the use of daily or prior-to-use quality control checks. In addition,
1228 appropriate instrument quality control should be established for background, voltage plateau,
1229 quenching, resolution and alpha-beta cross talk. Guidance on beta particle counting can be found
1230 in industry standards (ASTM D1890; D3648; E1329) and publications (NCRP Report 58; Knoll,
1231 1989; Lapp and Andrews, 1954; Price, 1989; USPHS, 1967).

1232 “Gross” alpha and beta counting of evaporated samples, wherein a multitude of alpha and beta-
1233 emitting radionuclides may exist, is typically used for screening of water samples. The
1234 application of such methods may be targeted for a specific radionuclide or a category of

1235 radionuclides such as the naturally occurring nuclides or a specific radionuclide in a facility
1236 effluent. However, extreme caution should be applied to the interpretation and use of such results
1237 without a full specific radionuclide characterization of the water source under investigation. The
1238 type of analysis is to be considered “gross” and, in most cases and for a variety of sound
1239 technical reasons, the gross measurement result does not equal the sum of the radionuclides
1240 contained in the sample.

1241 When specific radiochemistry is performed the beta-emitting radionuclide of interest will be
1242 isolated, concentrated and converted to a desired final chemical and physical form. Under these
1243 circumstances, the beta detection system should be calibrated for the radionuclide, chemical
1244 composition of the final sample form and the range of final sample weights expected from
1245 chemical recovery.

1246 15.10.2.2 Alpha Particle Interference and Beta Energy Resolution

1247 When properly operated or under optimal counting conditions (thin final samples or low LS
1248 quenching and high beta energy), most beta particle counting systems can separate alpha and beta
1249 particle detection events. However, the degree of alpha particle detection by the beta detector
1250 under consideration should be evaluated for each radionuclide, mixture of radionuclides or
1251 specific final sample form. Beta detection systems that are considered to have beta energy
1252 spectral resolution capabilities may be less affected by samples containing alpha-particle emitting
1253 radionuclides. However, for window gas proportional counters, alpha particle energy degradation
1254 by air, detector window or sample self absorption may lead to false beta detection without proper
1255 evaluation. A typical example would be a thick final sample matrix containing a mixture of alpha
1256 and low-energy beta-emitting nuclides.

1257 Some commercial window gas proportional counting systems have a feature for simultaneous
1258 alpha and beta particle counting that uses a voltage pulse height discrimination for the separation
1259 of beta and alpha particle detection events. A common and more historical means of separating
1260 alpha and beta particle events is to count the sample on the alpha proportional counting voltage
1261 plateau followed by a count on the beta (plus alpha) proportional counting voltage plateau. An
1262 alpha-to-beta crosstalk factor should be determine for the final sample weight and for the alpha
1263 and beta energies under consideration. The net beta count is determined by multiplying the alpha
1264 counts (from the alpha window for simultaneous counting or on the alpha counting plateau) by
1265 the alpha-to-beta cross talk factor.

1266 Window gas proportional counters typically are not used for beta spectrometers but instead
1267 record beta particle detection events giving rise a voltage pulse large than a discriminator setting.

1268 Under most circumstances, liquid scintillation counters have sufficient energy resolution
1269 capability and electronic discrimination to fully separate beta and alpha particle detection events.
1270 However, due to the nature of the beta energy continuum of an emission process and the inherent
1271 resolution of a liquid scintillation spectrometer, identification and quantification of multiple
1272 nuclides contained in the same sample is complicated unless their beta energies are widely
1273 separated. Computer software and beta interference factors should be applied in such cases.

1274 A liquid scintillation counter is typically used for Cerenkov counting. However, the final sample
1275 solution contains no scintillator as would a full liquid scintillation-sample cocktail. Cerenkov
1276 counting, due to the nature of measurement process, will not detect alpha particles of any energy
1277 or beta particles having an average beta energy less than 260 keV. Cerenkov counting is typically
1278 applied to single nuclide evaluations or for a mixture of two nuclides that have a differential
1279 maximum beta energies greater than 700 keV (e.g., ⁸⁹Sr and ⁹⁰Y). Beta interference factors should
1280 be applied in such cases.

1281 15.10.2.3 Liquid Scintillation Quenching

1282 The information on liquid scintillation quenching provided in Section 15.10.1.1 is applicable for
1283 beta particle detection. The degree of quenching should be determined for each radiochemical
1284 method, radionuclide or application. An appropriate correction factor/curve should be calculated
1285 and applied to the measurement results for the samples being evaluated. The magnitude of the
1286 quench correction may approach 50 percent in certain severe quenching situations.

1287 Cerenkov counting is less sensitive to “quenching” than liquid scintillation counters using
1288 scintillation cocktails. Typically, the final sample solution is a result of a control radiochemical
1289 process that eliminates most sources of contamination, chemical impurities and variability in the
1290 final sample solution.

1291 15.10.2.4 Beta Particle Attenuation

1292 Beta particle attenuation should be considered for window gas proportional, plastic scintillator
1293 and solid state detector counting applications. Beta particle attenuation can result from the
1294 interaction of a beta particle with the air, detector window or the matrix atoms of the final
1295 sample. Beta particle air attenuation is a function of the distance between the sample or source
1296 and the detector’s particle entrance window. Under most application for beta particle counting,
1297 this factor is typically insignificant compared to the other sources of beta particle attenuation.

1298 Figure 15.11 shows the attenuation of
 1299 beta particles is in air and water.
 1300 Consideration of the detector window
 1301 thickness and its beta particle
 1302 attenuation becomes important when
 1303 evaluating low energy beta particles
 1304 such as ^{14}C . Normally, the air and
 1305 detector window attenuation factors
 1306 are determined as a combined beta
 1307 attenuation-efficiency factor that
 1308 includes the sample self absorption for
 1309 a given application. In most
 1310 applications, a back scatter factor for
 1311 the material composition (Z value) of
 1312 the final sample mount is included
 1313 into a combined attenuation-
 1314 backscatter-efficiency factor or, more
 1315 simply, the combined detector
 1316 efficiency correction factor.

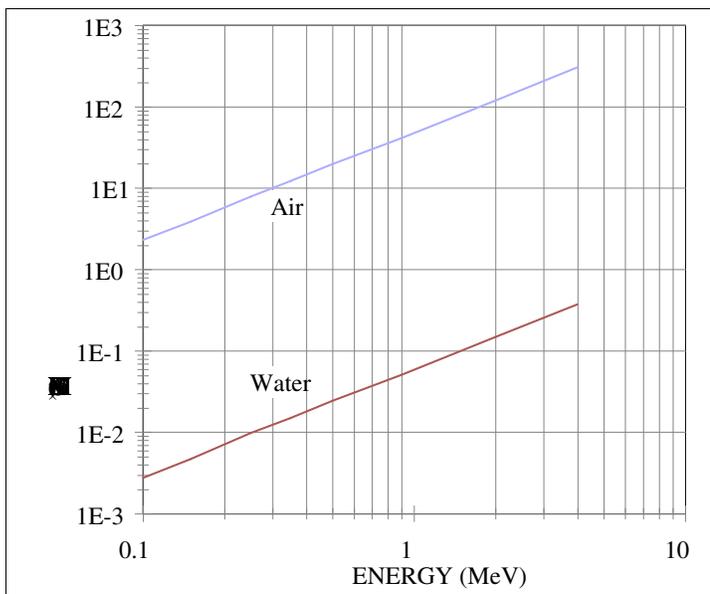


FIGURE 15.11 Range vs. Energy for Beta Particles in Air and Water

1317 For the lower to intermediate beta
 1318 particle energies, the combined detector
 1319 efficiency factor is a function of beta
 1320 energy, final sample mass and mass
 1321 composition. For beta particles having a
 1322 maximum beta energies greater than
 1323 1,500 keV, the combined detector
 1324 efficiency factor is nearly constant over
 1325 a final sample weight range of 0 to 5
 1326 mg/cm^2 . A typical combined beta
 1327 detector efficiency curve for ^{131}I (606
 1328 keV β_{max}) as CuI over a weight range of
 1329 0 to 50 mg is shown for a plastic
 1330 scintillator beta detector in Figure 15.12.
 1331 A complete review of the detection
 1332 method can be found in reference
 1333 (McCurdy et al., 1980).

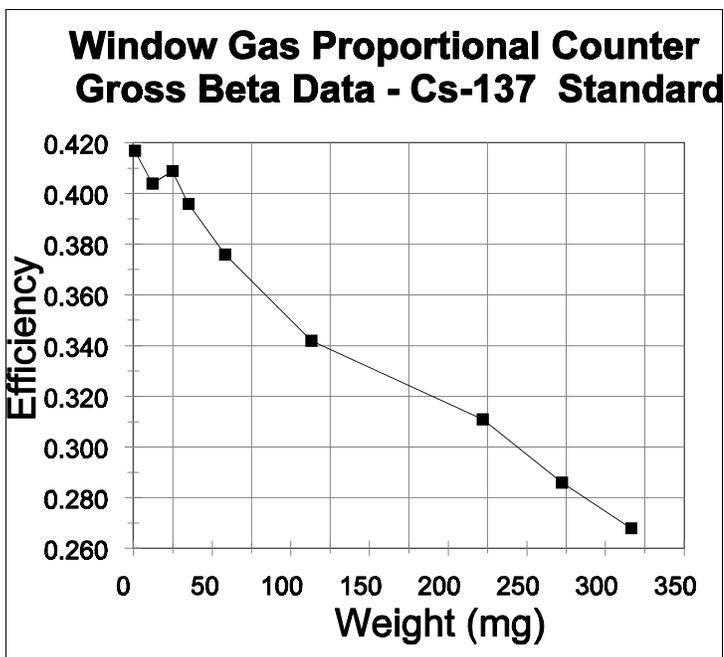


FIGURE 15.12 Beta Detector Efficiency Curve for ^{131}I vs. Weight

1334 15.10.2.5 Calibration

1335 Beta particle counting systems should be calibrated with the specific radionuclide under
1336 investigation or a surrogate radionuclide of similar beta energy having a comparable final sample
1337 composition and configuration. However, it should be mentioned that moderate to severe
1338 calibration biases may occur depending on the severity of the departure from the chemical
1339 composition of the final sample matrix and the beta energy of a surrogate. For this reason, the use
1340 of a surrogate radionuclide is discouraged unless the availability of the radionuclide of interest is
1341 non-existent. Corrections between the surrogate and radionuclide of interest should be
1342 determined and applied to sample results. For electroplated plated samples, a correction factor
1343 needs to be determined if the plating material of the surrogate is not the same as that used for the
1344 samples.

1345 Cerenkov counting normally involves a single radionuclide calibration (single energy calibration)
1346 for the final sample solution. Typically, the final sample solution is a result of a control
1347 radiochemical process that eliminates most sources of variability for the calibration process.

1348 Aqueous beta-emitting radionuclide calibration standards and sources are available from NIST or
1349 from a NIST-traceable commercial radioactive source manufacturers. The long-lived pure beta-
1350 emitting radionuclides available from NIST include: ^3H , ^{14}C , ^{63}Ni , ^{129}I , ^{89}Sr , ^{90}Sr , ^{99}Tc , ^{228}Ra , and
1351 ^{241}Pu . The majority of the gamma-emitting radionuclides also emit beta particles in the nuclear
1352 transformation process. Check Section 15.4 for the availability of known beta- gamma emitting
1353 radionuclides. Contact a NIST-traceable radioactive source manufacturer for the availability of
1354 other pure beta or beta/gamma-emitting radionuclides (ANSI N42.15, American National
1355 Standard Check Sources for and Verification of Liquid-Scintillation Counting Systems).

1356 Aqueous radioactive standards can be prepared in the appropriate geometry for LS or Cerenkov
1357 counting or through chemical processing precipitated or electroplated as final sample form for
1358 counting by a gas proportional, plastic or solid state beta detection system.

1359 15.10.2.6 Costs

1360 There are four principal beta detection methodologies available. Window gas proportional
1361 counting and liquid scintillation counting systems (Cerenkov counting as well) can be purchased
1362 with the option of readily available automatic sequential sample counting systems. Sample
1363 capacity is typically 100. These automatic sequential counting systems are available in the
1364 \$30,000 to \$50,000 range depending on options. Multiple detector window gas proportional
1365 counters having a simultaneous counting capability are available from some commercial

1366 manufacturers. The basic unit contains four detectors but several units can be combined to give
1367 eight or 16 detector systems. The basic price for such units is in the range of \$20,000 to \$50,000
1368 depending on the number of detectors and options.

1369 Solid state silicon surface barrier and ion implanted Ge or Si detectors are used to perform
1370 spectral analysis of beta emitting radionuclides. A solid state beta spectrometry system consists
1371 of a vacuum chamber, solid state detector, high voltage-preamp-amplifier instrumentation
1372 modules, a multichannel analyzer (MCA) or equivalent computerized MCA using an analog-to-
1373 digital converter and electronic data storage. Individual ion-implanted Ge detectors having an
1374 active area of 450-2,000 mm² and a 500 μm thickness range in price between \$1,300 and \$3,200.
1375 Beta resolution of these detectors is typically approximately 12 keV.0

1376 A beta spectrometry system consisting of eight detectors with vacuum pump and computer would
1377 be approximately \$30,000-\$40,000, without background reducing shielding. Solid state
1378 spectrometry systems for beta particle applications, unlike that for alpha particles, would be
1379 sensitive to external background from cosmic radiation, terrestrial radiation and inherent beta
1380 radioactivity in the surrounding materials.

1381 Automatic sample counting, plastic scintillator beta particle detection systems have not been
1382 commercialized for the radioassay laboratory setting. Most of these systems have been fabricated
1383 by the user from readily available components, electronic modules, multichannel analyzers and
1384 lead shielding. The cost of a single detector system is estimated to be less than \$15,000.

1385 Maintenance costs for the liquid scintillation counters, window gas proportional counters and
1386 alpha spectrometry systems have been discussed in Section 15.10.1.3.2 for alpha counting
1387 applications. If a laboratory already has existing units for the alpha particle measurement
1388 applications, there will be no additional maintenance cost relative to their use for beta particle
1389 measurements.

1390 There is no maintenance cost associated with the operation of a plastic scintillator beta
1391 spectrometry system.

1392 Costs associated with the maintenance of the room environment for the nuclear detection
1393 equipment should be considered. Service maintenance relative to the constant voltage supply or
1394 uninterruptable power sources as well as having a dust free constant temperature and humidity
1395 environment should be considered.

1396 15.10.2.7 Quality Control

1397 See section 15.10.1.4.

1398 **15.10.3 Gamma**

1399 15.10.3.1 Troubleshooting

1400 Once a gamma-ray spectrometry system has been established in accordance with the manufac-
1401 turer's or supplier's instructions, a daily count of a calibration or reference source should be
1402 performed to assure the system continues to operate properly. The three parameters that should
1403 be checked and recorded are: energy calibration (keV/channel), counting efficiency (count
1404 rate/decay rate), and gamma-ray peak resolution (FWHM). With the exception of a complete
1405 detector or electronic component failure (no pulses are detected at the preamp or multiplier
1406 phototube output), degradation of gamma-ray peak resolution will be the first indication that
1407 detector is not performing properly or that electronic noise has been introduced into the counting
1408 system by the preamplifier, amplifier, or multichannel analyzer. Any indications that the detector
1409 efficiency is not within statistical limits of expected values should be reported, since this value
1410 will be used to convert the observed count rate to decay rate. The energy calibration should either
1411 be recorded with sample spectral data or adjusted daily to a previously established constant
1412 value. This energy calibration should be accurately known so that nuclide identifications can be
1413 made. See page 51 for a list items to be checked if the counting system is out of specifications.

1414 Gamma-ray spectrometry systems are extremely sensitive to both electronic and environmental
1415 conditions. Temperature changes can cause spectral shifts and improper nuclide identifications
1416 because of incorrect energy calibrations. Excessive humidity in the detector preamplifier can
1417 cause high voltage arcing which results in poor peak resolution or complete system failure.
1418 Improper pole zero settings, which effects the shape of the pulses being analyzed, can cause
1419 degradation of peak shapes and resolution. Poorly conditioned NIM power can introduce
1420 electronic noise which will also result in degraded peak resolution. Routing of cables between the
1421 detector, electronics, multichannel analyzer, computers, and monitors is very important. The
1422 introduction of any spurious electronic noise into any of the components that make up the
1423 gamma-ray spectrometry system can degrade the resulting data.

1424 The need to make corrections for self-absorption in environmental samples during routine
1425 gamma-ray spectrometry cannot be overemphasized (Modupe et al., 1993). The correction to be
1426 made for the difference in self-absorption between calibration standards and sample matrices is
1427 usually small for intermediate and high energy photons, but it is not negligible at low energies

1428 where the photoelectric effect is the most important mode of attenuation. The photoelectric
 1429 process varies approximately as Z^{4-5} (Z is the atomic number of the elements in the medium) so
 1430 that a change in the elemental composition of a sample relative to a calibration standard can
 1431 require a correction factor for detector efficiency as high as a factor of 2.

$$I / I_0 = \frac{1 - e^{-\mu H}}{\mu H} \quad (15.1)$$

1432 The quantities μ and H are the linear attenuation coefficient and the thickness of sample,
 1433 respectively. I_0 and I are the intensities of the beam emerging from the sample container without
 1434 and with an absorbing matrix in place. This is the traditional self-absorption equation. For
 1435 complex counting geometries of homogeneous materials, an estimated average H (sample
 1436 thickness) can be used.

1437 The method for self-absorption correction at various energies requires that the linear attenuation
 1438 coefficient, μ , of the sample matrix be known. Knowledge of μ usually requires that the
 1439 elemental composition of the matrix be determined. The tedium and time required in elemental
 1440 analysis may make it impractical for routine gamma-ray analyses involving large numbers of
 1441 samples. Computer programs are available to calculate μ/ρ for various compounds when the
 1442 percent elemental composition of the compound is known. μ/ρ is computed as a linear
 1443 combination of the mass attenuation coefficients of the composite elements.

$$\mu / \rho = \sum (\mu_i / \rho_i) P_i \quad (15.2)$$

1444 where P_i is the percent by weight of the i th element in the compound.

1445 The gamma-ray path length, H , is equal to the thickness of sample. When performing gamma-ray
 1446 transmission measurements to determine μ a path length of H is used. To determine the self-
 1447 absorption correction for radioactive samples, the corrections are integrated for a path length of 0
 1448 to H .

1449 When a photon beam passes through a homogenous sample of mass attenuation coefficient, μ/ρ ,
 1450 density, ρ , and thickness, H , the percentage beam attenuation, A , is given by

$$A = \frac{I_0 - I}{I_0} 100\% = (1 - e^{-(\mu/\rho)Hp}) 100\% \quad (15.3)$$

1451 15.10.3.2 Calibration

1452 Most gamma-ray spectrometry systems are calibrated with either single or mixed standards in an
 1453 exact matrix and geometric form as the samples to be analyzed. However, there are computer
 1454 codes that can calculate detector efficiency from the physical dimensions of the detector and
 1455 sample counting geometry (Mitchell, 1986 and 1988, Hensley et al., 1997). Commercial
 1456 standards of single or mixed gamma-ray emitters in a matrix of known chemical composition and
 1457 density can be prepared in user supplied containers. Calibrations based upon these standards can
 1458 then be adjusted to correct for any differences in composition and density between the calibration
 1459 source and the sample (Modupe et al., 1993).

1460 Table 15.2 lists some gamma-ray emitting nuclides that can be used for energy and efficiency
 1461 calibration (Sanderson et al., 1993; Browne et al., 1986).

1462 **TABLE 15.2 Nuclides for Gamma-ray Spectrometer Calibration**

NUCLIDE	ENERGY (KeV)	HALF-LIFE
²¹⁰ Pb	46.5	22.3 years
²⁴¹ Am	59.5	432.2 years
¹⁰⁹ Cd	88.0	462.6 days
⁵⁷ Co	122.1	273 days
¹⁴¹ Ce	145.4	32.5 days
¹³⁹ Ce	165.9	137.7 days
²⁰³ Hg	279.2	46.6 days
⁵¹ Cr	320.1	27.7 days
¹¹³ Sn	391.7	115.1 days
⁸⁵ Sr	514.0	64.8 days
¹³⁷ Cs	661.7	30.0 years
⁵⁴ Mn	834.8	312.5 days
⁸⁸ Y	898.1, 1836.1	106.6 days
⁶⁵ Zn	1115.5	243.8 days
⁶⁰ Co	1173.2, 1332.5	5.27 years
⁴⁰ K	1460.8	1.28×10 ⁹ years

1480 15.10.3.3 Software

1481 Most laboratories are now using commercially available software for the analysis of gamma-ray
1482 spectra. These programs are easy to use and do not require the user to be an expert in gamma-ray
1483 spectrometry. An evaluation of some of these programs in 1987 indicated there were substantial
1484 differences in the abilities of the programs to resolve multiplets of unequal intensity and to
1485 analyze complex spectra (Sanderson 1988). Another evaluation was completed in 1992 (Decker
1486 and Sanderson, 1992) since many of the programs had undergone numerous revisions and there
1487 were a few new programs available. The second evaluation indicated a substantial improvement
1488 in the deconvolution of doublets and the results of the analysis of a Chernobyl air filter were
1489 much more consistent than when a similar filter was analyzed in the first evaluation. The six
1490 programs analyzed in 1991 include GAMMA-W from Germany, INTERGAMMA from France,
1491 OSQ/Plus from Canada, SAMP090 from Finland (supplied by Canberra Industries, USA) and
1492 OMNIGAM and GDR from the United States. Some of the features which contribute to a good
1493 program included the ability to display the spectrum as well as calculated calibration files, the
1494 ability to manually insert peaks during the fitting procedure, an extensive nuclide library and the
1495 ability to easily transfer nuclides to smaller, working libraries, an analysis report which includes
1496 the names of the calibration files used, a peak fit report including any problems with the shape of
1497 the peaks, and identification of the peaks used in the activity calculation as well as any problems
1498 with interfering lines.

1499 In 1996 the Environmental Measurements Laboratory of the U. S. Department of Energy began a
1500 Gamma Spectra Data Evaluation program (Decker et al., 1996) whose goal was to test the ability
1501 of the present day software to accurately identify and quantify the nuclides in a complex spectra
1502 and the ability of the user to properly utilize the software. In order to do this, synthetic spectra
1503 were generated using the computer code SYNTH developed by Walt Hensley at the Pacific
1504 Northwest National Laboratory. The spectra were then converted to a variety of formats on disk
1505 and Digital Equipment Corporation (DEC) TK 50 tape and sent to DOE laboratories and DOE
1506 contractors. A calibration spectrum, a background spectrum and three sample spectra were sent
1507 to each participant. These spectra simulated those that would be obtained when an air filter was
1508 counted 10 cm from a 22 percent coaxial detector with a 0.5 mm beryllium window. Two of the
1509 samples contained fallout and naturally occurring nuclides with half lives greater than thirty days.
1510 The third sample contained both short and long lived fission product nuclides. Thirty one
1511 laboratories participated using 16 different software packages. The software packages included
1512 Aptec, Vertechs GDR/P, Nuclear Data ASAP, various Ortec packages, and various Canberra
1513 packages for both the PC and the DEC MicroVax. Most of the laboratories did fairly well with
1514 the first two samples. A few laboratories reported nuclides that were not present in the third
1515 sample and did not accurately quantify those that were. The results did not seem to be software

1516 dependent but were due to the user utilizing or not utilizing available software features properly.
1517 There was quite a wide range of numbers for both the uncertainty terms and for the minimum
1518 detectable activities which seems to indicate we need a consistent way of calculating these terms
1519 to make them more meaningful.

1520 15.10.3.4 Costs

1521 Gamma-ray spectrometry systems can cost from \$14,000 to well over \$60,000 depending upon
1522 the choice of detector. For a 75 x 75 mm NaI(Tl) system the costs would be approximately
1523 \$1,000 for the detector, \$5,000 for a 10 cm graded lead shield, and \$8,000 to \$10,000 for a
1524 multichannel analyzer. Very large HPGe detectors will cost more than \$50,000. The actual
1525 detector cost will depend upon the size of the germanium crystal, its resolution, and method of
1526 cooling. Data reduction costs (software and computer) would be an additional expense for either
1527 type of system.

1528 NaI(Tl) detector systems do not require any additional maintenance beyond what any laboratory
1529 electronic system requires. Each HPGe detector will require approximately \$1,200 per year for
1530 liquid nitrogen to maintain their operating temperature. An electrical/mechanical cooler can be
1531 used in place of a liquid nitrogen cryostat but it will require 0.5 to 1.0 kW of power around the
1532 clock to operate. Both systems should be operated at constant temperature for reliable
1533 performance. This may require substantial air conditioning.

1534 15.10.3.5 Quality Control

1535 Initial data to prepare solid state gamma detector QC charts may be obtained by counting a mixed
1536 gamma point source between 20 to 30 times (Ideally, these counts should be over a period of
1537 several weeks. However, if time does not permit, the counts may be accumulated over 1 to
1538 7 days.) Two or three QC charts (depending on age of mixed gamma point source) are initially
1539 established for the mixed gamma point source and control limits are established for background.
1540 The three source charts cover the low energy (88 keV, ¹⁰⁹Cd), the medium energy (661.6 keV,
1541 ¹³⁷Cs), and the high energy (1,332.5 keV, ⁶⁰Co). The source is counted until between 10,000 to
1542 40,000 counts are obtained in each photopeak.

1543 Background QC charts are established according to the procedure already listed for Proportional
1544 and liquid scintillation counters with the following exception: the background is counted and the
1545 total counts in the spectrum are obtained by summing the counts in the entire spectrum.

1546 The resolution of the detector (FWHM) is measured each month and recorded, but it is not
1547 plotted. A NIST ^{60}Co source is positioned 25 centimeters from the end-cap face and counted for
1548 100 minutes. The FWHM is calculated by the peak search program for the 1,173.2 keV and
1549 1,332.5 keV peaks, and recorded in the logbook.

1550 When the energy of the source QC exceeds the specified energy tolerance (for example
1551 ± 0.75 keV) from its initial calibrated value, the analyzer system should be recalibrated. First
1552 determine whether a gain or zero shift has occurred. A gain shift is a nonlinear shift in channels
1553 of low and high energy peaks (i.e., ^{109}Cd peak shifts ± 1 channel and ^{60}Co peak shifts ± 3 channels).
1554 A zero shift is a linear shift in channels for both low and high energy peaks (ie., ^{109}Cd peak shifts
1555 ± 1 channel and ^{60}Co shifts ± 1 channel also). Make the appropriate adjustments to the amplifier
1556 (gain) or the Analog to Digital Converter (zero). Recalibrate the analyzer and record the slope
1557 (keV/channel) and the zero intercept in the log book. If the best fit of the recalibration curve is a
1558 nonlinear fit (quadratic), record the "Q" coefficient, keV/channel², in the log book. Also record
1559 the updated FWHM calibration factors, slope, offset, and FWHM at 1,332.5 keV in the log book.

1560 The following should be considered when QC checks are not within limits.

- 1561 • Is standard decay corrected to the proper date?
- 1562 • Check sample positioning.
- 1563 • Check for zero shift.
- 1564 • Check for gain shift.
- 1565 • Check full width at half-maximum.
- 1566 • Check nim bin power supply voltages (± 6 V, ± 12 V, ± 24 V).
- 1567 • Check efficiency tables.
- 1568 • Check for moisture on the detector due to recently filling the dewar with liquid N_2 .

1569 **15.10.4 Non-Nuclear Instrumentation**

1570 15.10.4.1 ICP-Mass Spectrometry

1571 ICP-MS is one of the most versatile and sensitive atomic spectroscopy techniques available. It
1572 can be used to determine the concentrations of over 70 elements. The detection limit of the
1573 technique extends down to the parts-per-billion range in soils and to the parts-per-trillion range in
1574 waters. This sensitivity makes ICP-MS an attractive complement to decay-counting techniques in
1575 the radiochemical analysis laboratory. For very long-lived radioisotopes (those with half-lives
1576 over 10,000 years, e.g., ^{244}Pu , ^{99}Tc , ^{129}I), ICP-MS may be faster and more sensitive than decay
1577 counting. In addition, sample preparation for ICP-MS can avoid some of the analyte separation

1578 and purification steps required for decay counting, providing an additional dimension of time
1579 savings. Another important feature of ICP-MS is its ability to provide isotopic distribution
1580 information (e.g., ^{238}U vs. ^{235}U). This information is frequently useful in determining the age
1581 and/or origin of materials. (ASTM C758, C759, C799)

1582 The isotopic discrimination capabilities of ICP-MS make possible the calibration technique
1583 known as isotope dilution. In this procedure, a sample is analyzed for one isotope after having
1584 been spiked with a different isotope of the same element (e.g., analysis of ^{235}U might involve
1585 spiking with ^{233}U). The spiked sample is carried through all preparation and analysis steps; in this
1586 way, any matrix or procedural effects that might influence the ^{235}U signal will influence the ^{234}U
1587 signal to precisely the same extent. Final quantization relies on measuring the ratio of unknown
1588 (here the ^{235}U signal) to the known (^{234}U) signal. Isotope dilution is a way of generating highly
1589 precise and accurate data from a mass spectrometer and has been used in the characterization of
1590 many certified reference materials.

1591 Although an ICP-MS instrument is extremely delicate, with proper care and preventive
1592 maintenance system up time should range between 80 to 95 percent. An initial investment of
1593 about \$200,000 will be required to obtain a current commercial state-of-the-art system. Annual
1594 maintenance costs will run from \$5,000 to \$20,000 depending on the purchase of a service
1595 contract.

1596 For more sophisticated measurements, at substantially higher cost, an ICP-MS with magnetic
1597 sector, instead of quadrupole, detection can be applied. Sector instruments are capable of
1598 resolving species of very similar mass. For example, ^{99}Tc might be resolved from a
1599 contamination of ^{99}Ru with a high-resolution mass spectrometric detector. More typically, high
1600 resolution instruments are employed for their higher signal/noise ratio, and therefore superior
1601 detection limits. A single-collector high-resolution ICP-MS can be purchased for roughly twice
1602 the cost of a quadrupole ICP-MS, or about \$300,000. For enhanced sample throughput a
1603 multiple-collector instrument might be purchased for about \$500,000. These instruments, like
1604 most analytical equipment, can be expected to require about 2 to 10 percent of their purchase
1605 costs in annual maintenance costs.

1606 Thermal ionization mass spectrometers are available at a cost of \$500,000. These instruments
1607 rely not on a plasma for ionization, but rather for thermal ionization from a heated filament. They
1608 provide more precise measurements than routine quadrupole ICP-MS but require substantially
1609 more delicate operator involvement, leading to markedly reduced sample throughput.

1610 Time-of-flight plasma mass spectrometers have just recently appeared on the market; they have
1611 not yet built up a historical record of performance that would permit reliable comparison with the
1612 ICP-MS equipment described above. Likewise, Fourier-transform mass spectrometers are still in
1613 the research phase and cannot yet be considered practical options for routine radiochemical
1614 analysis.

1615 15.10.4.2 Laser

1616 APPLICATION

1617 Lasers can be used to excite uranium (ASTM D5174) and lanthanide complexes in solution.
1618 During or following excitation, the complex relaxes to a lower energy state by emitting photons
1619 of light that can be detected. The amount of light produced is proportional to the uranium or
1620 lanthanide element concentration.

1621 The light emitted can be detected by fluorescence or phosphorescence. With fluorescence and
1622 phosphorescence, the detector is at right angles to Laser excitation. Fluorescence light is emitted
1623 simultaneous to the excitation.

1624 Phosphorescence detecting differs from fluorescence in that the light emitted is not simultaneous
1625 to the excitation. This enables the light source to be pulsed and the measurement to occur when
1626 the Laser source is off. This provides improved signal to noise over fluorescence. The light signal
1627 from organic material will decay promptly, since they have a short relatively lifetime, and not be
1628 available to the detector which is gated off at this initial time. A pulsed nitrogen dye Laser can be
1629 used as the source. Other Lasers can also be used. Chloride ion and other ions may cause
1630 interferences and may need to be removed before measurement.

1631 Kinetic phosphorimetry measures the rate of decay of the uranium or lanthanide element
1632 complex signal. Measurements are taken at fixed time intervals. In aqueous solution, the uranium
1633 or the lanthanide element is complexed to reduce quenching and increase the lifetime of the
1634 complex.

1635 UP/DOWN TIME

1636 Some reagents may have relatively short shelf life and need to be ordered accordingly. The life of
1637 a plasma cartridge is one to three years.

1638 COST

1639 The initial cost is about \$34,500 with a computer and \$53,500 with an automatic sample changer.
1640 The cost of replacing a plasma cartridge is \$1,400.00. The cartridge lifetime is 1 to 3 years
1641 depending on usage.

1642 15.10.4.3 Radionuclides Analyzed By Neutron Activation

1643 TECHNETIUM-99

1644 Neutron activation analysis methods have been employed since 1972 (Foti et al. 1972a; 1972b).
1645 The method was developed and applied for the analysis of ⁹⁹Tc in mixed fission products (Bate,
1646 1979).

1647 The method employs chemical separation of ⁹⁹Tc from most fission products by a cyclohexanone
1648 extraction from a basic carbonate solution. ⁹⁹Tc is stripped into water by addition of CCl₄ to the
1649 cyclohexanone phase and then adsorbed on an anion exchange column in a concentrated form.
1650 Neutron irradiation of the isolated ⁹⁹Tc could be made in the pneumatic facility at a high flux
1651 isotope reactor (e.g., at a flux of 5×10^{14} ng/cm²/sec for approximately 11 seconds. Thus, after
1652 irradiation ⁹⁹Tc is induced to ¹⁰⁰Tc, which, because of its 15.8 second half-life, requires an
1653 automatic process to measure its 540 and 591 keV gamma lines.

1654 The lower limit of detection of the analysis under these conditions is approximately 5 ng and
1655 samples up to 100 mL volume can be processed. The method has been applied successfully to
1656 reactor fuel solutions and off-gas traps containing 6.5×10^{-4} to 240 μg ⁹⁹Tc/mL.

1657 IODINE-129

1658 Iodine-129 can be determined by neutron activation and subsequent measurement of the
1659 12.4 hour ¹³⁰I produced by the neutron capture reaction. The method (Bate and Stokely, 1982)
1660 utilizes conventional I valence adjustments and solvent extraction to isolate the I fraction.
1661 Chemically separated ¹²⁹I is adsorbed on an anion exchange resin before being loaded for
1662 irradiation. With a neutron flux of 5×10^{14} ng/cm²/s for 100 seconds a lower limit of detection of
1663 0.03 ng can be achieved.

1664 ¹²⁹I also can be determined directly by mass spectrometry (Strebin et al., 1988). The measurement
1665 limit by this technique is approximately 2 femtograms.

1666 Special counting techniques have also
 1667 been applied to the analysis of ^{129}I .
 1668 Figure 15.13 shows an efficiency plot
 1669 using beta-gamma coincidence
 1670 counting.

1671 URANIUM, THORIUM, AND PLUTONIUM

1672 Neutron Activation analysis method was
 1673 employed to determine uranium in the
 1674 hydrogeochemical samples from
 1675 Savannah River Plants within the scope
 1676 of the National Uranium Resource
 1677 Evaluation Program sponsored by DOE.
 1678 Uranium was determined by cyclic
 1679 activation and delayed neutron counting
 1680 of the ^{235}U fission products. The method
 1681 relied on the absolute activation
 1682 techniques using the Savannah River Reactor Activation Facility.

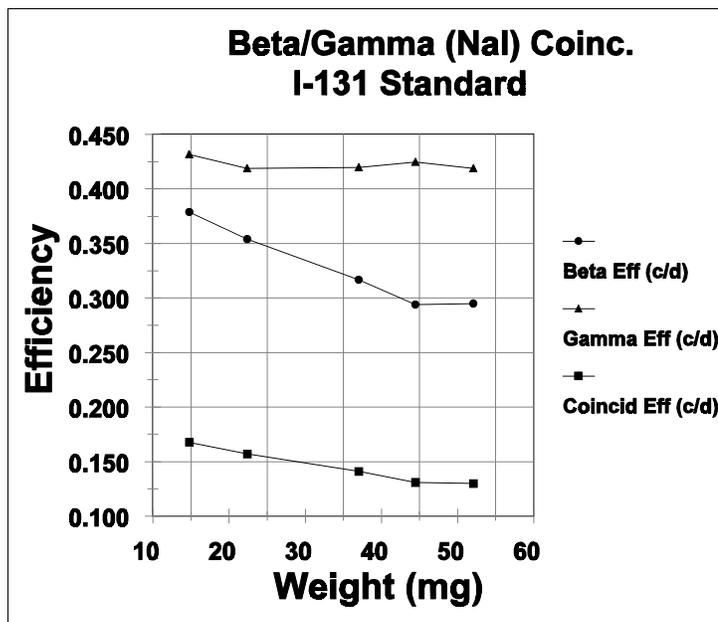


FIGURE 15.13 Beta-gamma coincidence efficiency curve for ^{129}I

1683 Neutron Activation Analysis followed by delayed-neutron detection was commonly used for
 1684 determination of ^{235}U , ^{239}Pu , and ^{232}Th (Echo and Turk, 1957; Hochel, 1979; Alfassi, 1990).

1685 15.11 References

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1875

ATTACHMENT 15A FIELD MEASUREMENTS

1876

15A.1 Introduction

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The scope of environmental remediation work taking place across the country in the public and private sector has led to a need to improve the speed and cost-effectiveness of measurements for characterizing contaminant levels at sites and assessing the results of cleanup efforts. In particular, the time for decisions that are required during soil excavation and waste segregation should be kept short to avoid delays that tend to increase labor costs. Thus, the time it takes to collect, prepare and analyze samples can be a limiting factor. To this end, one can use mobile laboratories at the field site to reduce sample handling and transit times. However, even with these, the sheer volume of samples can overwhelm processing and analytical capacity. Therefore, measurements performed directly in the field (in situ) that do not require the collection and processing of a sample are an attractive alternative. Fundamentally, a field measurement gives the concentration of a contaminant at the same place where one might otherwise have collected a sample. In effect, the instrument is brought to the sample rather than the sample to the instrument. Frequently, the field measurement can be performed within minutes with a result obtained in what is essentially “real time.”

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15A.2 Analytical Level of Measurements

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Over the years, field measurements have formed an important component of standard radiological surveys. Typically, these measurements have comprised scans for gross levels of alpha or beta/gamma radiation. These types of measurements, particularly where judgment is used to evaluate a change in an instrument or audible signal, are semi-quantitative in nature and therefore would be designated at analytical level 1 under the EPA classification system used in the past or Analytical Support Laboratory(ASL) level A of the American National Standards Institute (ANSI). These levels reflect the fact that the measurement is intended for screening purposes.

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However, field measurements can be performed at a higher analytical level. For example, an exposure rate measurement using a pressurized ionization chamber (PIC) is definitive for assessing the external dose rate from penetrating (gamma) radiation. In this situation, the PIC provides a direct reading of the desired measurement quantity at the actual point of interest.

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1904 Another example of a field measurement technique that has been used successfully since the
1905 1960s is in situ gamma-ray spectrometry (ICRU, 1994). This technique provides radionuclide-
1906 specific information. In its simplest application, a spectrometer could be used to identify
1907 characteristic peaks in the energy spectrum that would point to the presence of a particular
1908 radionuclide at the measurement location. On a semi-quantitative basis, *in situ* spectrometry
1909 could serve as screening technique where the relative count rates—in particular spectrum
1910 peaks—are compared among measurement locations. At a higher analytical level, an appropriate
1911 calibration can be performed so that a spectrometer could be used to determine the radionuclide
1912 concentration in the media under study. Since this represents a contaminant-specific
1913 measurement where particular QA/QC checks can be made, it would be classified traditionally at
1914 the data quality objectives (DQO) analytical level 2, or ASL B.

1915 Despite a number of successful applications of *in situ* spectrometry over the years, issues have
1916 arisen regarding the level of data quality that is obtained with this or any other field measurement
1917 technique for the purposes of demonstrating RCRA, CERCLA, and other regulatory compliance.
1918 In the past, field measurements by definition have not been considered to possess the quality
1919 control that needs to be established at a DQO analytical level of 4 (analogous to ASL D) in the
1920 laboratory. However, the distinction between screening level and higher level measurements is
1921 based on factors relating to data quality, which should be demonstrable. In principle, the rigorous
1922 QA/QC protocols and documentation required for analytical level 4, using EPA Contract
1923 Laboratory Program (CLP) procedures, or ANSI ASL D, could be applied to radionuclide-
1924 specific field measurements. Using field techniques at a higher analytical level is also in keeping
1925 with the latest EPA proposals for performance based measurement systems.

1926 Typically, a projection of cost or time savings using a novel field method leads to its substitution
1927 for a more standard sampling/laboratory analysis method. In doing so, the intended applications
1928 of the field measurement method need to be established clearly. Using the DQO process, the
1929 requisite analytical level can be determined for the data that are to be collected. This analytical
1930 level should then be demonstrated through an objective judgement process whereby the data
1931 quality indicators are critically examined. Included would be those that arise when applying the
1932 DQO process (the “PARCC” parameters: precision, accuracy, representativeness, completeness,
1933 and comparability). Other related indicators or elements which can be broken out separately and
1934 which need to be addressed include documentation, instrument operating conditions, site
1935 conditions, interferences, limitations, calibration procedures, minimum detectable concentrations,
1936 reference measurements, record keeping, quality improvement, and management assessment. The
1937 following sections will provide some discussion on each of these elements as they apply to field
1938 measurement data quality level. Although the discussion is based on experiences with in situ
1939 spectrometry, the elements would generally apply to other field measurement techniques as well.

1940 It would be expected that a demonstration of the data quality level of a field technique be
1941 performed in concert with regulatory bodies and stakeholders to obtain acceptance.

1942 **15A.3 Documentation of Methodology**

1943 A field measurement technique, like its counterpart in the laboratory, requires thorough
1944 documentation including the description of apparatus and materials, specification of personnel
1945 training/qualification level, listing of quality control checks, review of safety considerations, and
1946 issuance of non-conformance reports when necessary.

1947 Training materials, equipment manuals, reference texts, articles from technical journals, and
1948 laboratory reports are all potential sources of background information for describing a method. It
1949 would be expected that information be extracted from these sources and a comprehensive report
1950 issued that provides the necessary background and specifics for a particular site and application.
1951 This would essentially take the form of a written procedure. For multiple applications across a
1952 site, further detail may have to be provided in project-specific plans, as the conditions under
1953 which a technique is used may vary among areas. The guidance and recommendations given by
1954 standards groups can also form a key part of documentation. Adherence to these standard
1955 procedures allows one to proceed with some confidence in the measurements process. It is
1956 expected that standards groups will increasingly devote their efforts to field measurements
1957 techniques in the future.

1958 Individuals who will be working with the instruments and data collected need to be qualified.
1959 Educational backgrounds and necessary experience should be determined and appropriate
1960 training given for each area of work. Training and procedure manuals need to track revisions that
1961 invariably result as measurement programs progress.

1962 Quality systems documents would include a general site-wide quality plan with specific factors
1963 like performance tests, pre- and post-operational checks, frequency of calibrations, and replicate
1964 measurements addressed in a separate method-specific quality systems section or document.
1965 Quality systems includes documenting procurement specifications for apparatus and control of
1966 materials and services such as calibration sources. Also, the turnaround time for field
1967 measurements may be important to specify not only for cost and schedule control but for limiting
1968 the time lag between measurements under changing environmental conditions.

1969 Unforseen measurement conditions and unusual equipment malfunctions will lead to situations
1970 where doubt is cast on the validity of a field measurement. Tracking these failures will help to
1971 elucidate the problems over time and provide a basis for corrective actions and modifications to

1972 the procedures for future measurements. Situations where obvious bad data is collected despite
1973 the fulfillment of QC elements will require the writing and issuance of a non-conformance report
1974 with subsequent root cause analysis.

1975 **15A.4 Instrument Operating Conditions**

1976 Specification of instrument operating conditions is fundamental in the field as in the laboratory.
1977 These would include power and cooling requirements as well as an acceptable range in
1978 temperature and humidity conditions. The physical set-up of the instrument, such as a
1979 reproducible sample-detector geometry, also should be specified. For laboratory radioactivity
1980 counting systems, it is generally a planchet, can, bottle or similar small volume where the
1981 distribution of activity within the sample volume is assumed to be homogeneous. For a field
1982 measurement, the sample is in a form such as an area of ground, a storage drum, or wall.
1983 Distances and orientation to the measured area or object need to be specified and held within
1984 control limits.

1985 For a field measurement, the distribution of activity within the volume of measurement should be
1986 considered, since one does not usually have the luxury of mechanical blending as in the case of a
1987 laboratory sample. The field of view of the detector with respect to lateral and depth
1988 displacement within the volume under measurement needs to be established. For large volume
1989 sources, this generally means determining the response of the instrument across all angles or
1990 radiation incidence, not just the front face. While one cannot necessarily control the distribution
1991 of a contaminant, the instrument response needs to be established so that the integrated signal
1992 that it measures can be converted into a meaningful average result over the volume measured and
1993 the sensitivity to non-homogeneous activity distributions determined.

1994 **15A.5 Site Conditions/Limitations**

1995 Preparing a site for a field measurement is analogous to preparing a sample for analysis.
1996 Procedures need to be followed that will assure that the measurement will yield a valid result.
1997 This might include removing obstructions and accounting for topography and ground cover such
1998 as vegetation or surface water. The radiation absorption properties of the type of soil where
1999 measurements are made may have to be determined beforehand depending upon the energy and
2000 type of radiation being measured.

2001 A significant element which should be addressed in performing field measurements is changes in
2002 the "sample," i.e., changes in the field conditions at the measurement point. For example,
2003 measurements at the same location several days apart may be not be comparable if soil moisture

2004 conditions have changed. Precipitation events would increase soil moisture, while hot, dry
2005 conditions would lead to a decrease in soil moisture.

2006 Depending upon the instrumentation and the physical basis of the measurement, the effects of
2007 such variables as air and soil temperature, humidity, air pressure or related meteorological
2008 parameters may have to be taken into account.

2009 Limitations should be specified for a field measurement technique. They could include site
2010 conditions such as the water content of soil, the degree and type of ground cover, the size of an
2011 area, and the estimated depth of contamination. The radionuclide mix and the concentration level
2012 could also be limiting factors.

2013 **15A.6 Interferences**

2014 The effects of interferences need to be assessed for proper QC in a field measurement. As
2015 compared to a laboratory setting where there is generally a controlled environment, adverse
2016 instrument effects may result from extraneous signals or electronic noise that could be produced
2017 by power line or other electromagnetic interference. Interferences in a measurement could also
2018 result from personnel—whether instrument operators or other workers—who enter into a
2019 measurement area and attenuate the measured radiation.

2020 Whereas a laboratory counting system may employ a shield to block out background radiation, a
2021 field measurement system is exposed to ambient radiation. If significant direct or scattered
2022 (shine) radiation is present from extraneous sources, collimation or shadow shielding may be
2023 necessary. In high radiation fields, the effects of ionization in electronic components may present
2024 a problem. In this case the sensor assembly could be kept at the measurement point with the
2025 signal processing and other electronics kept at a distance.

2026 As in the case of laboratory analysis, attention should be given to the mix of radionuclides that
2027 may be present. Interferences can result from the inability to resolve the primary energies emitted
2028 by the nuclides or because there is a high amount of secondary (scattered) radiation present.

2029 **15A.7 Calibration**

2030 Calibration requires that the instrument response to a known level of measured substance be
2031 determined. This generally takes the form of measuring standard reference materials or samples
2032 spiked with known quantities.

2033 Direct calibrations using standards or spikes are usually applied to laboratory-based counting
2034 systems since only small quantities are needed for the sample volumes used. For field
2035 measurements, direct calibrations using large volume sources with a known concentration can be
2036 performed as well, although this is generally impractical and potentially expensive. In place of
2037 this, a field calibration factor for a particular source geometry and matrix composition can be
2038 derived using a two-step process. This entails determining the response to incident radiation
2039 (fluence) as a function of energy and angle (by experimental and/or theoretical means) and then
2040 calculating the fluence at the point of measurement from a given source geometry and matrix.
2041 Two-step calibration methods sometimes are applied to laboratory sample counting geometries as
2042 well.

2043 Although a two-step process may be used for field calibrations, traceability still can exist insofar
2044 as certified point or other sources can be used in the calibration process. The calibration factor
2045 may actually represent an integrated response to a collection of sources or a single source at
2046 many different positions. In the case of a spectrometer, calibration points will need to be spaced
2047 out across the energy range of interest. Depending upon saturation effects, the calibration may
2048 also have to extend across a range in concentrations to assess the effects of signal processing
2049 dead time and pulse pile-up.

2050 **15A.8 Minimum Detectable Concentrations**

2051 Standard to any high quality measurement technique and integral to the DQO process is an a
2052 priori estimate of the detection limits of the measurement system. This needs to be done for a
2053 field measurement technique, although it may be necessary to first obtain preliminary readings in
2054 the area where measurements are to be performed. For example, the minimum detectable
2055 concentration (MDC) for a particular radionuclide will be affected by the continuum of scattered
2056 radiation present in a spectrum from other radionuclides in the soil or from sources of scattered
2057 radiation outside the area under investigation.

2058 In some situations the sensitivity for a given count time can actually be higher for a field
2059 measurement as compared to a laboratory-based sample measurement, thus producing a lower
2060 MDC. This will result when the field detector gives a higher count rate per unit concentration
2061 because there is a far larger sample being analyzed.

2062 **15A.9 Precision**

2063 The precision of field measurements is determined with replicate measurements as in the case of
2064 laboratory measurements. To avoid potential changes in field conditions, replicate measurements
2065 can be performed sequentially with minimum time lag.

2066 In many cases, a field measurement is a non-destructive technique. Thus, replicate measurements
2067 are easily performed. Using the results from a successive set of measurements (5 to 10), a
2068 standard deviation about the mean can be calculated. This can then be compared to the counting
2069 error for a single measurement that is based on Poisson statistics to assess precision.

2070 Rather than perform many replicate measurements at one point, it can be more instructive to
2071 perform two or more measurements at several different points. The reproducibility can thus be
2072 judged for a variety of site conditions.

2073 **15A.10 Accuracy**

2074 Estimates of accuracy for a field measurement can be obtained through uncertainty propagation
2075 just as in the laboratory. Factors to consider include potential bias due to uncertainties in the
2076 calibration source, variations in the assumed sample/detector geometry, uncertainties in the
2077 sample matrix composition, environmental conditions, as well as the statistical counting error.

2078 Overall system accuracy can be checked with comparisons to other techniques, or to results from
2079 an independent organization using the same technique.

2080 **15A.11 Representativeness**

2081 Representativeness refers to the degree to which a measurement reflects the condition at a
2082 location or whether a group of measurements reflects the conditions in a particular area.
2083 Generally, one desires that measurements (or samples) provide a value of a radionuclide
2084 concentration that in turn yields the best dose estimate (and thus risk) to a member of a critical
2085 group for a particular scenario. In order to achieve representativeness, a number of samples or
2086 measurements in a given area would be required in order to achieve a given confidence level or
2087 power using a statistical test.

2088 Representativeness is affected by the heterogeneity of the contaminants in the media under
2089 investigation. Perhaps more than any other factor, field and laboratory measurements may differ
2090 at any particular measurement location due to the effects of heterogeneity. Heterogeneity can

2091 exist in both the lateral and depth distribution of a contaminant and can take the form of changes
2092 in concentration across various distances: a centimeter or less, as would result from hot particles;
2093 meters, as might occur from dumping and localized spills; and tens or hundreds of meters, as
2094 from up-wind airborne sources. Survey designs incorporate techniques and sample/measurement
2095 densities to accommodate these variations. The number of measurements and the standard
2096 deviation about the mean are fundamental parameters to judge whether the mean concentration
2097 that is measured is within a certain confidence limit. These parameters can be used to compute
2098 the t statistic or applied to other statistical tests.

2099 Where variations in concentration occur on a scale of tens of meters or more, it can be expected
2100 that either field measurements or soil sampling will give similar results. It is where the variations
2101 on the scale of a few meters or less occur that agreement between any particular pair of field
2102 measurement and soil sample results might suffer. However, if the mean concentration in an area
2103 should be determined, a sufficient number of measurements or samples can ultimately yield the
2104 same average result, regardless of where the measurements or samples are taken within the area
2105 under investigation.

2106 Depending upon the objectives of a measurement program, a field method could inherently have
2107 an advantage over discrete sampling. If the viewing area of a field instrument is significantly
2108 larger than the area of a soil sample, a set of field measurement results would tend to show a
2109 smaller standard deviation as compared to a set of soil sample data in a heterogeneous area. The
2110 mean obtained for a given number of measurements would then be more representative of the
2111 true mean. A wide measurement area represented by a field method could also be consistent with
2112 the assumptions of a dose model which averages over a large area.

2113 **15A.12 Completeness**

2114 Measurement losses can occur in the field just as sample losses can occur in the lab. They result
2115 from equipment failure, improper measurement procedures, or environmental factors beyond the
2116 control of operators. Survey designs should incorporate allowances for sample losses by
2117 specifying the collection of more than just the minimum number of samples needed to support a
2118 decision.

2119 There is somewhat of an advantage for a field technique in that QC checks can be performed at
2120 the time of the measurement. Problems can then be immediately identified and the data rejected
2121 on the spot. Another measurement can then be performed in place of the lost measurement.

2122 **15A.13 Comparability**

2123 Comparability is a critical factor that readily establishes the validity of a field technique. It can be
2124 established by performing a study in which field measurement results are compared to those
2125 given by an independent technique, such as sampling and laboratory analysis. In some situations,
2126 it may be possible to compare two different field techniques.

2127 In performing a direct comparison study, it is important to establish that the two techniques are
2128 measuring the same thing. For instance, a technique that measures a contaminant concentration in
2129 the surface soil may compare poorly to one that is integrating down to greater depths. This
2130 situation would result where there is a non-uniform concentration depth profile of the
2131 contaminant. Where comparisons are made to soil samples, core depths can be adjusted to better
2132 match the effective viewing depth of the field measurement. The lateral distribution of the
2133 contaminant concentration across the ground could also be a factor. In this situation, compositing
2134 samples may be required to yield a better average with which to compare a field technique.

2135 Other factors to consider for data comparability include the soil moisture and stone content of
2136 soil. Where contaminant concentrations are determined with a field technique, the value is based
2137 on the wet weight of the soil in contrast to laboratory analysis which is performed on a dry
2138 weight basis. Corrections to one of the data sets therefore need to be applied. Similarly, one
2139 should consider the effects of soil sample preparation where large stones are screened out. The
2140 concentration which is then determined is based on the activity associated with the finer particle
2141 content of the soil. If there is little or no activity in the coarser fraction, a concentration for a soil
2142 sample would be higher than that given by a field technique which has averaged in the stone
2143 content.

2144 In place of comparing single field measurement points to single or composited samples, one can
2145 instead compare the averages of sets of field measurements to sets of soil samples over a
2146 particular size area. This would be useful to establish comparability where there is a known
2147 heterogenous distribution of the contaminant and the techniques under comparison are measuring
2148 very different areas of soil.

2149 **15A.14 Reference Measurements**

2150 An important QC practice in the laboratory involves the regular analysis of reference materials to
2151 confirm system calibration and performance. In practice, an analogous check can be performed
2152 for measurements in the field. A reference measurement location at a site can be designated as a
2153 field quality control station where routine, perhaps daily, measurements of the contaminants of

2154 concern are performed. QC charts can be kept which show the results of these measurements and
2155 control limits can be specified accordingly. Unusual trends can then be identified early and
2156 corrective actions taken before unusable data is generated. Measurements at a station such as this
2157 also serve to demonstrate the effects of environmental variables such as temperature and
2158 humidity.

2159 To further qualify a field station, intensive sampling can be performed with laboratory analyses
2160 to determine contaminant concentrations. In this situation, relatively homogeneous conditions
2161 (soil type, contaminant concentration) would make the comparison more favorable and help to
2162 trace any bias between measurement methods that might be observed.

2163 In addition to reference materials, the analysis of blanks is a regular feature of laboratory-based
2164 counting systems. This establishes that contamination of equipment and materials has not
2165 occurred. Similar contamination can occur to field instrumentation such as wind blown soil
2166 particles in crevices, encrusted mud on the underside of equipment, or soil plugs in tripod legs.
2167 For a field measurement technique, it may be possible to check self-contamination by performing
2168 measurements in a background area where the contaminant in the soil is essentially zero. If the
2169 contaminant is present in background, such as ¹³⁷Cs from nuclear weapons fallout, an offsite area
2170 at least can serve to establish a regional baseline measurement. As a standard measure of
2171 precaution, routine scanning of equipment can be performed with friskers, especially after work
2172 in highly contaminated areas.

2173 **15A.15 Record Keeping**

2174 Field personnel need to use log sheets or books to record necessary information about the site
2175 conditions, measurement parameters, and data storage. In place of chain of custody forms for
2176 samples, analogous records may be required for data printouts or electronic files of results
2177 (spectral data) obtained in the field as they pass through different levels in the organization (data
2178 entry, data analysis, validation, etc.).

2179 Maintenance logs or files on specific pieces of equipment need to be kept. Factory repairs or in-
2180 house replacement of components should be noted as any changes to an instrument are likely to
2181 require recalibration. Equipment and component failures should also be tracked.

2182 **15A.16 Quality Improvement**

2183 Operating experience generally leads to fuller knowledge of instrument performance and
2184 characteristics as well as better recognition of precursors to problems. Based on control chart

2185 records, observations and correlations with other factors associated with a measurement,
2186 breakdown and repair logs, and the information contained in any non-conformance report,
2187 procedures can be modified to improve data recovery and usability. In time, the net effect of
2188 changes incorporated in standard operating procedures will lead to improvements in performance
2189 tests. Along with the identification of limiting factors and the development of solutions, it may
2190 be possible to justify raising the analytical level of the measurement based on the quality control
2191 indicators.

2192 **15A.17 Management Assessment**

2193 In addition to the quality control elements in place when a field technique is demonstrated
2194 initially, systems need to be in place to insure that data quality is maintained in subsequent
2195 measurements once the technique is used routinely. Deployment of a field methodology on a
2196 broader scope generally entails use by non-experts, i.e., individuals not associated with the
2197 development or implementation of an instrument. For this reason, internal assessments may be
2198 needed in the form of independent oversight (audits). The data verification and validation process
2199 can be used to insure the fulfillment of QC checks. Ultimately, data may have to be reviewed and
2200 approved by individuals who have expertise with the measurement system.

2201 **15A.18 Combined Laboratory and Field Measurements**

2202 Laboratory and field measurement techniques are not mutually exclusive. They can frequently be
2203 used in concert to achieve better and more cost-effective radiological surveys. A likely
2204 combination would be reliance on field methods which are faster with the laboratory method
2205 serving as a QC check. Appropriate ratios in the number of field to lab measurements would have
2206 to be established based on expert judgement and by reviewing the data quality objectives. The
2207 ratio could vary from area to area within a site depending upon the situation and the presence of
2208 complicating factors.

2209 **15A.19 References**

2210 International Commission on Radiation Units and Measurements (ICRU), 1994. *Gamma-Ray*
2211 *Spectrometry in the Environment*, Report 53, Bethesda, MD.