Designation:D 4962–95ᵋ¹ Designation: D 4962 – 02 (Reapproved 2009)

Standard Practice for
NaI(Tl) Gamma-Ray Spectrometry of Water[1]

1. Scope

1.1 This practice covers the measurement of gamma-ray emitting radionuclides in water by means of gamma-ray spectrometry. It is applicable to nuclides emitting gamma-rays with energies greater than 50 keV. For typical counting systems and sample types, activity levels of about 40 Bq (1080 pCi) are easily measured and sensitivities of about 0.4 Bq (11 pCi) are found for many nuclides (11-10). [2] Count rates in excess of 2000 counts per second should be avoided because of electronic limitations. High count rate samples can be accommodated by dilution or by increasing the sample to detector distance.

1.2 This practice can be used for either quantitative or relative determinations. In tracer work, the results may be expressed by comparison with an initial concentration of a given nuclide which is taken as 100 %. For radioassay, the results may be expressed in terms of known nuclidic standards for the radionuclides known to be present. In addition to the quantitative measurement of gamma radioactivity,gamma-ray activity, gamma-ray spectrometry can be used for the identification of specific gamma-ray emitters in a mixture of radionuclides. General information on radioactivity and the measurement of radiation has been published (11 and 12). Information on specific application of gamma-ray spectrometry is also available in the literature (3(13-16).

1.3

1.3 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

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

2. Referenced Documents

2.1 *ASTM Standards:*[3]

D 3648 Practices for Measurement of Radioactivity Practices for the Measurement of Radioactivity

D 4962 Practice for NaI(Tl) Gamma-Ray Spectrometry of Water

E 181 Test Methods for Detector Calibration and Analysis of RadionuclidesRadionuclides

3. Summary of Practice

3.1 Gamma-ray spectra are commonly measured with modular equipment consisting of a detector, an analyzer, a memory amplifier, multi-channel analyzer device, and a permanent storage device. computer (17 and 18).

3.2 Thallium-activated sodium-iodide crystals, NaI(Tl), which can be operated at ambient temperatures, are often used as gamma-ray detectors in spectrometer systems. However, their energy resolution limits their use to the analysis of single nuclides or simple mixtures of a few nuclides. Resolution of about 7 % (45 keV full width at one half the Cs-137 ¹³⁷Cs peak height) at 662 keV can be expected for a NaI(Tl) detector in a 7576 mm by 7576 mm-configuration.

3.3 Interaction of a gamma-ray with the atoms in a NaI(Tl) detector results in light photons that can be detected by a multiplier phototube. The output from the multiplier phototube and its preamplifier is directly proportional to the energy deposited by the incident gamma-ray. These current pulses are fed into an amplifier of sufficient gain to produce voltage output pulses in the amplitude range from 0 to 10 V.

3.4 A multichannel pulse-height analyzer is used to determine the amplitude of each pulse originating in the detector, and accumulates in a memory the number of pulses in each amplitude band (or channel) in a given counting time. Computerized systems with stored programs and interface hardware can accomplish the same functions as hardwired multichannel analyzers. The

primary advantages of the computerized system include the capability of programming the multichannel analyzer functions and the ability to immediately perform data reduction calculations using the spectral data stored in the computer memory or mass storage device time (417 and 18).). For a 0 to 2 MeV spectrum two hundred data points are adequate.

3.5 The distribution of the amplitudes (pulse heights) of the pulse energies, represented by the pulse height, can be separated into two principal components. One of these components has a nearly Gaussian distribution and is the result of total absorption of the gamma-ray energy in the detector; this peak is normally referred to as the full-energy peak or photopeak. The other component is a continuous one, lower in energy than that of the photopeak; thisphotopeak. This continuous curve is referred to as the Compton continuum and results from interactions wherein the gamma photons lose only part of their energy to the detector. Other peaks components, such as escape peaks, backscattered gamma-rays, or x-rays from shields, are often superimposed on the Compton continuum. These portions of the curve are shown in Fig. 1 and Fig. 2. Escape peaks will be present when gamma-rays with energies greater than 1.02 MeV are emitted from the sample (5(19-24). The positron formed in pair production is usually annihilated in the detector and one or both of the 511 keV annihilation quanta may escape from the detector without interaction. This condition will cause single- or double-escape peaks at energies of 0.511 or 1.022 MeV less than the photopeak energy. In the plot of pulse height versus count rate, the size and location of the photopeak on the pulse height axis is proportional to the number and energy of the incident photons, and is the basis for the quantitative and qualitative application of the spectrometer. The Compton continuum serves no useful quantitative purpose in photopeak analysis and must be subtracted when from the photopeak to obtain the correct number of counts before peaks are analyzed.

3.6 If the analysis is being directed and monitored by an online computer program, the analysis period may be terminated by prerequisites incorporated in the program. If the analysis is being performed with a modern multichannel analyzer, analysis Analysis may also be terminated when a preselected time or total counts in a region of interest or in a specified channel is reached. Visual inspection of a cathode-ray tube (CRT) display of accumulated data the computer monitor can also be used as a criterion for manually terminating the analysis on either type of data acquisition systems. analysis.

3.7 Upon completion of the analysis, the spectral data are interpreted and reduced to nuclide activity of becquerels (disintegrations per second) or related units suited to the particular application. At this time, the spectral data may be inspected on the CRTmonitor to identify the gamma-ray emitters present. This is accomplished by reading the channel number from the x-axis and converting to gamma-ray energy by means of an equation relating channel number and gamma-ray energy. If the system is calibrated for 10 keV per channel with channel zero representing 0 keV, the energy can be immediately calculated. In some
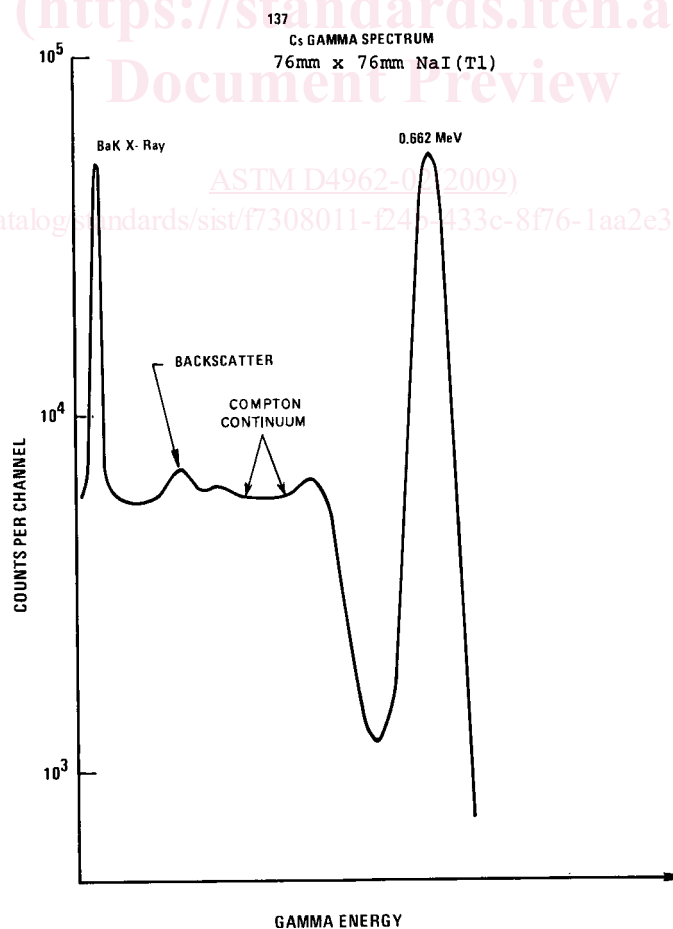
FIG. 1 Compton Continuum

**Part of ²⁴Na Gamma
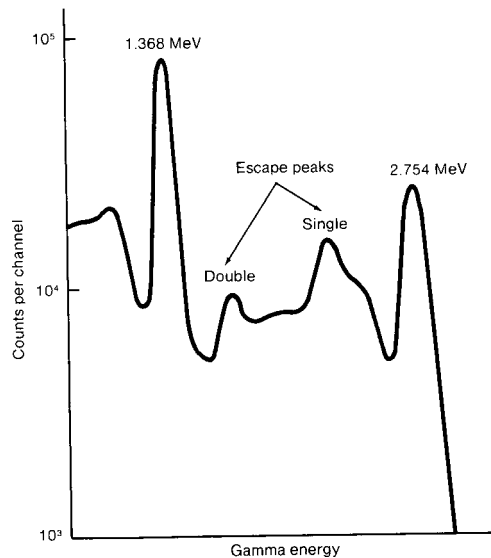Spectrum 76mm × 76mm NaI(Ti)**

FIG. 2 Single and Double Escape Peaks

systems the channel number or gamma-ray energy in keV can be displayed on the ~~CRT~~monitor for any selected channel. Identification of nuclides may be aided by ~~catalogs~~libraries of gamma-ray spectra and other nuclear data tabulations **(~~6~~25-30).**

~~3.7.1Computer programs for data reduction have been used extensively although calculations for some applications can be performed effectively with the aid of a desktop or pocket calculator (1~~

3.7.1   Data reduction of spectra involving mixtures of nuclides is usually accomplished using a library of standard spectra of the individual nuclides acquired under conditions identical to that of the unknown sample **(25-30).** ~~Data reduction of spectra involving mixtures of nuclides is usually accomplished using a library of standard spectra of the individual nuclides acquired under conditions identical to that of the unknown sample (6).~~

4. Significance and Use

4.1   Gamma-ray spectrometry is used to identify radionuclides and to make quantitative measurements. Use of a computer and a library of standard spectra will be required for quantitative analysis of complex mixtures of nuclides.

4.2   Variation of the physical geometry of the sample and its relationship with the detector will produce both qualitative and quantitative variations in the gamma-ray spectrum. To adequately account for these geometry effects, calibrations are designed to duplicate all conditions including source-to-detector distance, sample shape and size, and sample matrix encountered when samples are measured. This means that a complete set of library standards may be required for each geometry and sample to detector distance combination that will be used.

4.3   Since some spectrometry systems are calibrated at many discrete distances from the detector, a wide range of activity levels can be measured on the same detector. For high-level samples, extremely low efficiency geometries may be used. Quantitative measurements can be made accurately and precisely when high activity level samples are placed at distances of 1 m or more from the detector.

4.4   Electronic problems, such as erroneous deadtime correction, loss of resolution, and random summing, may be avoided by keeping the gross count rate below ~~100000~~2 000 counts per ~~minute~~second and also keeping the deadtime of the analyzer below 5 %. Total counting time is governed by the activity of the sample, the detector source distance, and the acceptable Poisson counting uncertainty.

5. Interferences

5.1   In complex mixtures of gamma-ray emitters, the degree of interference of one nuclide in the determination of another is governed by several factors. If the gamma-ray emission rates from different radionuclides are similar, interference will occur when the photopeaks are not completely resolved and overlap. A method of predicting the gamma-ray resolution of a detector is given in the literature ~~(7~~**(31).** If the nuclides are present in the mixture in unequal portions radiometrically, and nuclides of higher gamma-ray energies are predominant, there are serious interferences with the interpretation of minor, less energetic gamma-ray photopeaks. The complexity of the analysis method is due to the resolution of these interferences and, thus, one of the main reasons for computerized systems.

5.2   Cascade summing may occur when nuclides that decay by a gamma-ray cascade are analyzed. Cobalt-60 is an example; 1172 and 1333 keV gamma-rays from the same decay may enter the detector to produce a sum peak at 2505 keV and cause the

loss of counts from the other two peaks. Cascade summing may be reduced by increasing the source to detector distance. Summing is more significant if a well-type detector is ~~used.~~used.

5.3 Random summing occurs in all measurements but is a function of count rate. The total random summing rate is proportional to the square of the total number of counts. For most systems, random summing losses can be held to less than 1 % by limiting the total counting rate to ~~1000~~2 000 counts per second (see ~~General~~ Methods E 181).

5.4 The density of the sample is another factor that can affect quantitative results. This source of error can be avoided by preparing the standards for calibration in matrices of the same density of the sample under analysis.

6. Apparatus

6.1 *Gamma Ray Spectrometer*, consisting of the following components, as shown in Fig. 3:

6.1.1 *Detector Assembly*—Sodium iodide crystal, activated with about 0.1 % thallium iodide, cylindrical, with or without an inner sample well, 51 to 102 mm in diameter, 44 to 102 mm high, and hermetically sealed in an opaque container with a transparent window. The crystal should contain less than 5 µg/g of potassium, and should be free of other radioactive materials. In order to establish freedom from other radioactive materials, the manufacturer should supply the gamma-ray spectrum of the background of the crystal between 80 and 3000 keV. The crystal should be attached and optically coupled to a ~~multiplier phototube.~~ photomultiplier. (The ~~multiplier phototube~~photomultiplier requires a preamplifier or a cathode follower compatible with the amplifier). The resolution (FWHM) of the assembly for the photopeak of cesium-137 should be less than 9 %.

6.1.2 *Shield*—The detector assembly shall be surrounded by an external radiation shield made of massive metal, equivalent to 102 mm of lead in gamma-ray attenuation capability. It is desirable that the inner walls of the shield be at least 127 mm distant from the detector surfaces to reduce backscatter. If the shield is made of lead or a lead liner, the shield must have a graded inner shield of 1.6 mm of cadmium or tin lined with 0.4 mm of copper, to attenuate lead x-rays at 88 keV, on the surface near the detector. The shield must have a door or port for inserting and removing samples.

6.1.3 *High Voltage Power/Bias Supply*—High-voltage power supply of range (usually from 500 to 3000 V and up to 10 mA) sufficient to operate a NaI(Tl) detector, ~~multiplier phototube,~~photomultiplier, and its preamplifier assembly. The power supply shall be regulated to 0.1 % with a ripple of not more than 0.01 %. Line noise caused by other equipment shall be removed with radiofrequency filters and additional regulators.

6.1.4 *Amplifier*—An amplifier compatible with the preamplifier or emitter follower and with the pulse-height analyzer.

6.1.5 *Data Acquisition and Storage Equipment*—A multichannel pulse-height analyzer (MCA) or stand-alone analog-to-digital-converter (ADC) under software control of a separate computer, performs many functions required for gamma-ray spectrometry. An MCA or computer collects the data, provides a visual display, and outputs final results or raw data for later analysis. The four major components of an MCA are the ADC, the memory, the control, and the input/output circuitry and devices. The ADC digitizes
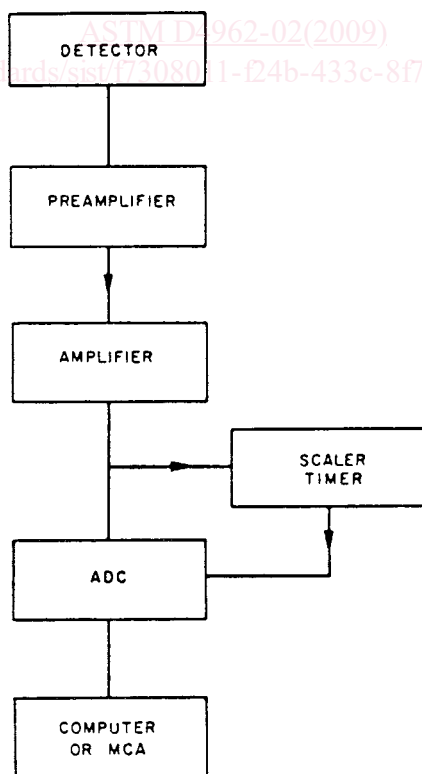

FIG. 3 Gamma Spectrometry System

the analog pulses from the detector amplifier. These pulses represent energy. The digital result selects a memory location (channel number) which is used to store the number of events which have occurred with that energy. Simple data analysis and control of the MCA is accomplished with microprocessors, which control factors such as the input/output, channel summing over set regions of interest, and system energy calibration.

6.1.6 *Data Storage*— Because of the use of microprocessors, modern MCAs provide a wide range of input and output (I/O) capabilities. Typically, these include the ability to transfer any section of data to one or more of the following: ~~terminal, line~~ printer, ~~cassette tape,~~ floppy or hard disk, x-y plotter, and computer interfaces by way of a serial, parallel, ethernet or ~~parallel~~USB port.

7. Container for Test Specimen

7.1 Sample mounts and containers must have a convenient reproducible geometry. Considerations include commercial availability, ease of use and disposal, and the containment of radioactivity for protection of the working environment and personnel from contamination. The evaporation of liquid samples to dryness is not necessary and liquid samples up to several ~~liters~~litres may be used. However, samples that have been evaporated to dryness for gross beta counting can also be used for gamma-ray spectrometry. Massive samples may cause significant self-absorption of low-energy gamma-rays and degrade the higher-energy gamma-rays. Therefore, it is important to calibrate the detector with standards of the same geometry and density. A beta absorber consisting of about 6 mm of aluminum, beryllium, or plastic may be used for samples that have a significant beta activity and high beta energies.

8. Single or Simple Mixtures of Radionuclides

8.1 *Calibration and Standardization* :

8.1.1 Begin operation of the instrumentation and detector according to the manufacturer's instructions. Initial set-up includes all electronic adjustment to provide constant operating conditions consistent with the application and life expectancy of the calibrations. Adjust the analog-to-digital converter gain and threshold, amplifier gain and zero-level, and detector high voltage, or bias, to yield an optimum energy calibration, usually 10 keV per channel. Modern commercial equipment is capable of linearity to the extent that the energy may be interpreted by the operator directly to the nearest 5 keV simply by reading the channel number of the highest channel in a peak. The energy calibration is usually accomplished with radioactive sources covering the entire range of interest. All efficiency calibrations and source analyses are performed with the same gain settings and the same high voltage setting. Obtain efficiency calibrations by placing an appropriate volume of an NIST traceable radionuclide standard solution containing 100 to 10 000 Bq in a container and placing the container on the detector or in the detector well.

8.1.2 *Preparation of Apparatus*:

8.1.2.1 Follow the manufacturer's instructions, limitations, and cautions for the set up of and the preliminary testing for all of the spectrometry equipment to be used in the analysis. This equipment could include detectors, power supplies, preamplifiers, amplifiers, multichannel analyzers, and computing systems.

8.1.2.2 Place an appropriate volume of an NIST traceable standard or an NIST traceable mixed standard of radionuclides in a sealed container and place the container at a desirable and reproducible source-to-detector distance. The solution above should provide about 100 counts per second in the peaks ~~of interest and be made up of standard sources traceable to a nationally certified laboratory.~~ interest. In all radionuclide measurements, the volumes, shape, and physical and chemical characteristics of the samples, and the corresponding standards, ~~and their containers~~ must be as identical as practicable for the most accurate results. If precipitates or residues are to be analyzed, then evaporate the standards on the same type of mount as the sample.

8.1.3 *Energy Calibration*:

8.1.3.1 Multichannel analyzers and NaI(Tl) detectors being produced today are capable of producing an almost linear energy response.

8.1.3.2 The energy calibration (channel number of the multichannel analyzer versus the gamma-ray energy) of the detector system is accomplished at a fixed gain using standards containing known radionuclides. The standards should be in sealed containers and should emit at least four different gamma-ray energies covering the range of interest, usually 50 keV to 2000 keV in order to test for system linearity. Some commercially available nuclides suitable for energy calibration are: ~~lead-210,~~ [210]Pb, 46.5 keV; ~~americium-241,~~ [241]Am, 59.5 keV; ~~cadmium-109,~~ [109]Cd, 88 keV; ~~cerium-141,~~ [141]Ce, 145 keV; ~~chromium-51,~~ [51]Cr, 320 keV, ~~cesium-137,~~ [137]Cs, 662 keV, ~~manganese-54,~~ [54]Mn, 835 keV; ~~sodium-22,~~ [22]Na, 511 and 1275 keV; ~~yttrium-88,~~ [88]Y, 898 and 1836 keV; ~~cobalt-60,~~ [60]Co, 1173 and 1332 keV; equilibrated ~~radium-226,~~ [226]Ra, 186, 352, 609, 1120, and 1765 keV; others have been listed **(417 and 18).** Verify the ~~radionuclidie~~radionuclide purity of the standards periodically to ensure against accidental contamination or the presence of long lived impurities by comparing the observed spectra with the spectra published in the literature **(417 and 18).**

8.1.3.3 Calibrate the multichannel analyzer to cover the range of interest. If the range is 50 to 2000 keV, adjust the gain of the system until the ~~cesium-137~~ [137]Cs photopeak, 662 keV, is about one-third full scale. Leaving the gain constant, locate at least three other photopeaks of different energies, covering the same range. ~~Determine and record~~ Calibrate the ~~multichannel analyzer channel number corresponding to~~ system based upon the ~~maximum count rate for each~~ manufacturer's instructions ~~of the four gamma-ray energies. Using rectangular coordinate paper, plot the gamma-ray energy versus the channel number for each of the major gamma-ray energies.~~ at least two, and preferably three, photopeaks. A near linear relationship will be observed if the equipment is operating properly. Samples should not be analyzed if there is a significant non-linear relationship. ~~Calculate the slope and~~

intercept of the line using a least squares calculation. If the spectrometry system is computerized, follow the appropriate manufacturer input instructions for the determination of the slope and intercept. During each day in which the spectrometry system is being used to analyze samples, repeat the above sequence of operations using at least two different gamma-ray energies. If the slope and intercept are essentially unchanged, the energy calibration data remain valid. If an appreciable change in the slope or intercept is evident, rerun the entire calibration procedure. For low-level samples counted over the course of several days, monitor the position of two peaks, for example, the 511 keV annihilation peak and ^{40}K (1460 keV). If these remain constant the system is assumed to be operating properly.

8.1.4 *Photon Detection Efficiency Calibration*:

8.1.4.1 Accumulate an energy spectrum using sealed, calibrated radioactive standards in a desired and reproducible counting geometry (see sSection 8.1.2.2). Accumulate at least 10 000 net counts (total counts minus the eCompton continuum and ambient background) in each full-energy gamma-ray peak of interest. Compare the live time of the count to the half-life of the radionuclide of interest. If the live time is a significant portion of the half-life, apply a correction factor for decay during the count **(625-30)**.).

8.1.4.2 Correct the radioactivity standard source gamma-ray emission rate for the decay from the time of standardization to the time at which the count rate is measured.

8.1.4.3 Calculate the full-energy peak efficiency, *E*, using Eq 1:

$$E = P/G \tag{1}$$

where:
E = full-energy peak efficiency (counts per gamma-ray emitted),
P = net gamma-ray count in the full energy peak of interest (counts per second), and
G = gamma-ray emission rate (gamma-rays per second).

If the standard source is calibrated as to activity, calculate the gamma-ray emission rate using Eq 2:

$$G = A \times P \tag{2}$$

where:
A = number of nuclear decays per second, and
P = probability per nuclear decay for the gamma-ray.

8.1.4.3 Calculate the full-energy peak efficiency using the observed net count rate in the full energy peak and the gamma-ray emission rate of the standard source. The gamma-ray emission rate being the product of the known nuclide activity and fraction of decays producing the respective full-energy emission. Practice D 4962 presents equations for this purpose in sections 11 and 41.

8.1.4.4 Many modern spectrometry systems are computerized and the determination of the gamma-ray efficiencies are determined automatically at the end of an appropriate counting interval. Refer to the manufacturer's instructions for specific input requirements.

8.1.4.5 Plot the values for the full-energy peak efficiency (as determined in 8.1.3) versus gamma-ray energy. The plot will allow the determination of efficiencies at energies for which standards are not available and to show that the algorithms used in computerized systems are providing valid efficiency calibrations. A typical plot is shown in Fig. 4.

8.1.4.6 Once the efficiencies have been determined, it is unnecessary to recalculate them unless there is a change in resolution, geometry, or system configuration. However, count a standard of a long-lived nuclide such as Cs-137 periodically as a quality control check. (See Section 13.)

9. Complex Mixtures of Radionuclides

9.1 Because of the inherent energy resolution of NaI(Tl) detectors, gamma-ray peaks in complex mixtures of nuclides may not be separated sufficiently for quantification as outlined in Section 8. It may not even be possible to visually locate individual peaks if their energies are similar or their intensity is too low in relation to other gamma-rays present in the spectrum. Complex mixtures of as many as 10 to 20 radionuclides can be quantified mathematically with computer programs using a linear least squares technique (1). (1-10).

9.1.1 When using these techniques care should be given to the following recommendations:

9.1.1.1 *System Gain and Zero Energy Channel*—Monitor and record the exact gain and zero energy channel of the spectrometer. If the computer analysis program performs gain or base line (zero energy channel) corrections, or both, on sample data, then the library of standards data must be obtained under uniform and precise calibration conditions.

9.1.1.2 *Library Standards*—The least squares analysis technique is a linear combination of all of the data contained in the standards library. Therefore, the standards library must contain a spectra of every component in the sample; in addition, these spectra must be obtained from the purest radionuclides available.

9.1.1.3 *Counting of Library Standards*:

(a) (a) All gamma-ray spectra will contain a background component. The activity of the library standards must be high enough so that this background component will be insignificant even though all computer programs make some kind of a background correction.

(b) (b) The duration of the counting period for the standard library spectra should be long enough to obtain statistically valid