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Guidelines for Gamma-Ray Spectroscopy Measurements of ^{235}U Enrichment

L.A. Kull and R.O. Ginaven

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TECHNICAL SUPPORT ORGANIZATION

BROOKHAVEN NATIONAL LABORATORY, UPTON, NEW YORK 11973

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GUIDELINES FOR GAMMA-RAY SPECTROSCOPY
MEASUREMENTS OF ^{235}U ENRICHMENT

by

L. A. Kull* and R. O. Ginaven*

Under Supervision of The
Technical Support Organization

*La Jolla Research & Business Associates
P. O. Box 1454, 1200 Prospect Street
La Jolla, California 92037

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Brookhaven National Laboratory
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GUIDELINES FOR GAMMA-RAY SPECTROSCOPY

MEASUREMENTS OF ^{235}U ENRICHMENT

1. Introduction

It is intended that this guideline provide a convenient description of the enrichment measurement technique, for determining the general applicability of the technique to a given sample or class of samples, and for estimating the level of measurement accuracy obtainable for a given set of sample characteristics. The guideline also includes an introduction to the instrumentation required for these measurements, and, it is hoped, will provide some aid in the specification of key instrumentation characteristics.

1.1 Description of the Technique

The alpha decay of ^{235}U to ^{231}Th is accompanied by the emission of a prominent gamma ray at 185.7 keV (4.3×10^4 of these 185.7 keV gamma rays are emitted per second per gram of ^{235}U). Due to the relatively low energy of these gamma-rays, those generated within a sample of modest size are strongly attenuated before reaching the surface so that the gamma-ray flux emitted by a thick* sample approximates that of an infinitely thick medium. As will be shown, the 185.7 keV activity from a semi-infinite thick sample of uranium bearing material is directly proportional to the ^{235}U enrichment and, under certain circumstances, is approximately independent of the material composition. A measurement of this 185.7 keV activity with a suitable detector therefore forms the basis for an enrichment measurement technique. A simple analytical expression describing this enrichment measurement is developed below.

Figure 1 illustrates the basic measurement configuration. The sample is assumed to contain a homogeneous distribution of uranium as well as other non-nuclear matrix materials. The measured activity $dC(x)$ (counts/sec) in the full energy peak of a detector spectrum resulting from detection of the unscattered flux of 185.7 keV gamma rays from a thin slab of

*The samples are thick compared to several mean free paths of a 185.7 keV gamma ray in the sample material (see Section 2.1 for further discussion).

Fig. 1. Detector configuration for enrichment measurements.

material with thickness dx (cm), area A (cm^2) located a distance x (cm) from the interior surface of the container (wall thickness, d) is given by

$$dC(x) = E \rho_u A \frac{\Omega}{4\pi} a \epsilon \exp[(-\rho_u \mu_u - \sum_i \rho_i \mu_i)x] \exp(-\rho_c \mu_c d) dx \quad (1)$$

where

E = enrichment of the uranium, $^{235}\text{U}/\text{U}$ (≤ 1)

ρ_u, ρ_i, ρ_c = density of the uranium, matrix material (i), and container wall (g/cm^3)

μ_u, μ_i, μ_c = mass attenuation coefficient* for 185.7 keV gamma rays in uranium, matrix material (i), and container wall (cm^2/g)

a = specific 185.7 keV gamma-ray activity of ^{235}U (4.3×10^4 gamma rays/sec.g)

ϵ = absolute detector full energy peak efficiency for detecting 185.7 keV gamma rays (≤ 1)

$\frac{\Omega}{4\pi}$ = fractional solid angle subtended by the detector ($\Omega \leq 2\pi$) (See Section 3 for numerical calculations of this factor.)

Equation (1) is correct if all gamma-ray trajectories are parallel to the collimator axis and is a close approximation if the collimator is long compared to its diameter. The expression is useful for analyzing less ideal geometries. Actual instruments must be calibrated empirically.

The combined count rate from all the material viewed by the detector from $x = 0$ to $x = L$ is therefore given by:

$$C = \int_0^L dC(x) = E \rho_u A \frac{\Omega}{4\pi} a \epsilon \exp(-\rho_c \mu_c d) \int_0^L \exp\left(-\rho_u \mu_u - \sum_i \rho_i \mu_i\right)x$$

*See Appendix 1 for a brief discussion of gamma ray attenuation coefficients.

If length L is taken to be a depth within the sample such that most of the 185.7 keV gamma rays generated at that depth are absorbed before reaching the surface, then:

$$\exp(-\rho_u \mu_u - \sum_i \rho_i \mu_i) L \ll 1 \quad (2)$$

Performing the integration and using the condition given by Eq. (2) results in

$$C = E \left(\frac{a}{\mu_u} \right) \frac{\epsilon A \Omega}{4\pi} \left(1 + \sum_i \frac{\rho_i \mu_i}{\rho_u \mu_u} \right)^{-1} \frac{\exp(-\rho_c \mu_c d)}{\rho_c \mu_c} \quad (3)$$

physical constants
detector + collimator
sample material composition
container wall effect

This analytical expression has been separated into several individual factors in order to aid in identifying those quantities which affect the measured count rate. Clearly, the count rate is proportional to the enrichment (E), but in addition, is affected by several other sample characteristics as well.

The term "enrichment meter" has often been applied to this technique for applications where the container wall effect is either small or is constant over a range of samples and where the term related to composition of the matrix is approximately equal to one, i.e.,

$$\left(1 + \sum_i \frac{\rho_i \mu_i}{\rho_u \mu_u} \right) \approx 1 \quad (4)$$

Examples for which the above approximation holds include pure uranium metal where the above term is equal to 1.000,

and UO_2 where it is equal to 1.012. Count rates measured under identical conditions from suitable thick samples of uranium metal and UO_2 in the same type of container can therefore be directly interpreted in terms of uranium enrichment, with only a small bias introduced by the difference in the sample matrix composition. This approximation works best for matrix materials of low atomic number.

The reason for this can be understood from the average behavior of the attenuation coefficients as a function of atomic number (Z) as shown in Fig. 2. Here one observes that materials with $Z \leq 30$ (which include many common matrix materials such as paper, glass, carbon, fluorine, iron, etc.) have approximately equal attenuation constants ($\mu \sim 0.13 \text{ cm}^2/\text{g}$). Rewriting Eq. 4 using the formalism defined in Appendix 1, we obtain

$$\left(1 + \frac{\mu_M \rho_M}{\mu_u \rho_u} \right) \approx 1 \quad .$$

Inserting the average value for μ_M ($Z \leq 30$) and μ_u ,

$$\left[1 + .07 \left(\frac{\rho_M}{\rho_u} \right) \right] \approx 1 \quad .$$

For many samples, the uranium density (ρ_u) is much greater than the density of the matrix (ρ_M) so that the approximation given by Eq. 4 obviously holds for a large number of common fuel cycle samples. More will be said concerning the effects of different matrix materials in Section 2.

The description above outlines the basic concept underlying an enrichment measurement using the 185.7 keV gamma-ray from ^{235}U . The remainder of this guideline discusses the important considerations which must be taken into account before applying this technique to any of the broad spectrum of available nuclear material samples. Wherever possible, mathematical expressions are given in this guide which will aid the user in determining the practicality of applying this measurement tech-

Fig. 2. Gamma ray attenuation coefficient (μ) for $E_\gamma = 186 \text{ keV}$ versus atomic number (Z). [Extrapolated from data contained in Photon Cross Sections, Attenuation Coefficients, and Energy Absorption Coefficients from 10 keV to 100 GeV, N SRDS-NBS 29 (1969). Ref. 11]

nique to specific samples. Numerical results are worked out for some common sample types and where appropriate, actual measured results are also included.

1.2 Basic Equipment Requirements

A block diagram showing the fundamental components of the measurement system is shown in Fig. 3. Both thallium-doped sodium iodide (NaI(Tl)) scintillation detectors and solid state lithium-drifted germanium (Ge(Li)) detectors have been used to perform these measurements. The system(s) shown represent conventional gamma-ray spectroscopy systems that are presently available commercially in modular or single unit construction. Data from a Ge(Li) detector has usually been stored in a multichannel analyzer (System B, Fig. 3) while the NaI detectors have been used with both multiple single-channel analyzers or multichannel analyzers (Systems A or B, Fig. 3). Both these different data storage and related data analysis options are discussed in Section 4.

An introduction to the use of scintillation and solid state gamma-ray detectors for nuclear material measurements is given in Ref. 1. Detector and associated electronic specifications, which are important for ^{235}U enrichment measurements, are given in Section 3. Good descriptions of enrichment measurement systems which have been used in nuclear fuel cycle plants are found in Refs. 2 through 8. Measurement results from several of these systems are summarized in Section 5.

A. Multiple Single Channel System

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Fig. 3. Simplified block diagram of an enrichment measurement system.

2. Factors Affecting Measurement Accuracies

Certain sample characteristics and measurement practices can affect the 185.7 keV activity observed from uranium samples. If these are not controlled or properly accounted for in determining the enrichment from the measured activity, errors will be introduced into the enrichment results. This section identifies important sources of possible measurement error.

2.1 Sample Thickness Effects

In order that Eq. 3 be applicable for a given sample, it is necessary that the thickness* of the sample be comparable to that amount of material required to produce strong attenuation of 185.7 keV gamma rays. To determine whether a sample meets this criterion, it is useful to compare the actual thickness of the sample with a characteristic length x_0 , where x_0 is defined to be that thickness of material which produces 99% of the measured 185.7 keV activity.

$$x_0 = - \frac{\ln(.01)}{u'}$$

$$x_0 = \frac{4.61}{u'}$$

(5)

where

$$u' = u_u \rho_u + \sum_i u_i \rho_i$$

Calculated values of x_0 for several common materials are given in Table 1.

*Thickness here refers to the linear dimension of the sample which is parallel to the detector collimator axis; this direction is labeled (x) in Fig. 1.

Table 1

Calculated Values of the Thickness of Material Required
to Produce 99% of the Measured 185.7 keV
Gamma-Ray Activity for Common Fuel Cycle Materials

Material	Density (g/cm ³)(Ref.9)	x_o (cm)(Ref.10)	x_o (in.)(Ref.10)
U(metal)	18.7	0.17	0.067
UF ₆	4.7	0.94	0.37
UO ₂	10.9	0.32	0.13
U ₃ O ₈	7.3	0.49	0.19
Uranyl Nitrate	2.8	2.0	0.79

To a very good approximation, the measured activity is independent of the sample thickness for samples thicker than x_o . Conversely, measurements of samples which are significantly thinner than x_o are subject to important inaccuracies introduced by varying sample thickness. As an example of an application of the results from Eq. 5 and Table 1, consider a 0.95 cm diameter (excluding cladding material) fuel pin containing UO₂. Results from Table 1 show that if the pin is scanned along its length with a closely collimated detector, no significant inaccuracies will be introduced due to reasonable fluctuations in the diameter of the fuel, since the nominal thickness (0.95 cm) is about three times deeper than that depth of material (given by x_o) responsible for producing 99% of the measured activity.

Several general observations can be made from these results:

- A large number of uranium samples common to fuel cycle plants will easily qualify as thick samples.

- For thick samples, the measured enrichment corresponds only to the surface layers of the sample material.
- If the technique is used on thin samples, Eq. 3 is not applicable and the measured accuracy of the results will be affected by variations in sample thickness. Appendix 1 contains analytical expressions for calculating the effect of variations in sample thickness on the measured activity.

2.2 Sample Inhomogeneity Effects

Inhomogeneities in matrix material composition, uranium density, and uranium enrichment within the measured volume of sample material (as characterized by the depth x , Section 2.1) will all produce changes in the measured 185.7 keV activity and hence will affect the accuracy of an enrichment calculated on the basis of that activity. Although it is impossible to determine the exact magnitude of changes in the measured activity produced by sample inhomogeneities, several general conclusions have been drawn from simple analytical models.

- There is a small to negligible effect on the measurement accuracy due to variations in content of low atomic number ($Z < 30$) matrix materials within the measured volume. However, care should be exercised in applying this technique to samples containing high atomic number matrix materials ($Z > 50$) or low concentrations of uranium. In these two cases, variations in material content can become an important limitation on measurement accuracy. A rough indicator which can aid in identifying samples for which inhomogeneities in matrix composition may be important is:

$$\sum_i \frac{\mu_i \rho_i}{\mu_u \rho_u} > 0.1 \quad (6)$$

Inhomogeneities in uranium density within the measured volume will also produce small to negligible effects on the accuracy of the results when the samples contain low atomic number matrix materials. However, as in the case of inhomogeneities in matrix composition, uranium density inhomogeneities can limit the accuracy of the measurement for samples containing higher atomic number matrix materials or low concentrations of uranium. The same inequality used above can again help to identify possible "problem" samples, i.e.,

$$\sum \frac{\mu_i \rho_i}{\mu_u \rho_u} > 0.1 \quad (6)$$

The most significant inaccuracies can arise in enrichment measurements when the uranium enrichment varies throughout the sample. In these cases, there is obviously no reason to expect that the measured volume near the surface adequately represents the average enrichment of the entire sample volume. Extreme care should be exercised in interpreting the results when this technique is applied to samples which are known to contain inhomogeneous distributions of uranium bearing materials with different enrichments.

2.3 Container Wall Effects

Variations in the thicknesses of the sample container walls can significantly affect the accuracy of this technique. The percent change in the measured activity ($\Delta C/C$) due to a small change Δd in the container wall thickness can be expressed as follows:

$$\frac{\Delta C}{C} (\%) = \mu_c \rho_c \Delta d \times 100 \quad (7)$$

Calculated values of $\Delta C/C$ corresponding to a change in container thickness Δd of .0025 cm (.001 in.) for common container materials are given below in Table 2.

Table 2

Calculated Values of the Change in Measured Sample Activity (%) Per .0025 cm (.001 in.) Change in Container Wall Thickness for Common Container Materials

Material	Density ^(Ref.9) (g/cm ³)	$(\frac{\Delta C}{C})$ (%) for $\Delta d = .0025$ cm ^(Ref.10)
Steel	7.8	.3
Aluminum	2.7	.09
Polyethylene	0.95	.04

As an example of the use of Eq. 7 and Table 2, consider a set of uranium samples held in steel containers fabricated with a wall thickness specified to within $\pm .025$ cm. The measured activity for a given enrichment can be expected to vary within $\pm .025(\text{cm}) \times .3$ (% per .0025 cm) = $\pm 3.0\%$ due to variations in wall thickness for this set of samples.

In some cases, unknown sample wall thicknesses can be measured using an ultrasonic technique and a simple correction can be applied to the data to account for attenuation of the 185.7 gamma rays passing through the sample wall (see Eq. 3). Commercial equipment is available to measure wall thicknesses ranging from about 0.025 to 5.1 cm (0.01 to 2.0 in.) to relative accuracies of 1.0% to 0.1% respectively. In order to make this correction, the composition of the container wall must be known and the container wall surface should be relatively clean and flat. General conclusions regarding the effect of the container wall thickness on enrichment measurements are given below.

- If possible, the same type of container should be used for the standard and the unknown samples. Otherwise, a calculated or measured correction must be applied to the measured data to take into account the difference in container wall thickness and composition.
- From Table 2 it is evident that variations in thicknesses of light, low atomic number container materials (e.g., polyethylene) have less effect on the accuracy of the measurement than materials with higher atomic number such as steel.
- Close control over container wall specifications can increase the accuracy of the measurement if preliminary analyses identify variations in wall thickness as an important source of error.
- The effect of variations in container thickness over an individual container may be minimized by averaging several measurements made at different locations or by rotating or scanning the sample during the measurement.
- If necessary, the effect of varying wall thickness can easily be determined experimentally by measuring the variations in the net 185.7 keV count rate from a ^{235}U source placed in a representative sampling of empty sample containers. Any ^{235}U source can be used, provided the geometry of source and detector is held constant throughout the series of measurements.

2.4 Background Radiation Effects

Three sources of background radiation can contribute to events recorded by the detector within the full energy peak width of the 185.7 keV line. These are:

1. A Compton continuum from higher energy gamma rays emitted from radioactive materials within the sample.

2. Overlapping of spectral peaks from gamma-ray lines which lie close in energy to the 185.7 keV line. These gamma rays are emitted by materials other than ^{235}U which are also contained within the sample.
3. Ambient backgrounds produced by natural causes and by other radioactive materials located in the vicinity of the measurement system.

2.4.1 Compton Continuum

The 185.7 keV peak from ^{235}U is always superimposed on a continuous and relatively smooth background from Compton scattered higher energy gamma rays. For many samples this background is predominantly produced by the 765 and 1,001 keV gamma rays of $^{234\text{m}}\text{Pa}$, a daughter of ^{238}U . (See Fig. 4.) The ^{234}Th and $^{234\text{m}}\text{Pa}$ daughter products of ^{238}U are generally removed when uranium is chemically processed. After processing they grow back in at a rate determined by the 24.1 day half-life of ^{234}Th . Since in most cases this Compton background behaves smoothly in the vicinity of the 185.7 keV peak, it can be readily subtracted by any of several methods (see Section 5), leaving only the net counts in the 185.7 keV full energy peak. It is essential that this continuum be subtracted from the peak, since its magnitude can vary considerably depending on the composition, age and enrichment of a sample.

It is also important to note that under certain conditions the shape of the background continuum can also change from sample to sample. These changes can be produced by: (1) radioactive species (other than uranium) in the sample that emit higher energy gamma rays or (2) different geometrical distributions of $^{234\text{m}}\text{Pa}$ within a sample. An example of such interference is given in Ref. 8. Spurious results were obtained on some 2.5 ton cylinders of low enriched UF_6 due to a radioactive daughter of uranium which had plated out on the cylinder wall. Ge(Li) systems are less affected by changes in the shape of the continuum since the excellent energy resolution of these detectors requires that only a narrow band of the continuum be sampled in order to make a background correction.

COUNTS PER CHANNEL (RELATIVE)

Fig. 4. Gamma ray pulse height spectra from uranium oxide with natural (0.7%) ^{235}U enrichment.

A) 3 in x 3 in NaI detector

B) 50 cc Ge(Li) detector

2.4.2 Overlapping Peaks

Backgrounds produced by interfering gamma-rays which lie close in energy to the 185.7 keV line pose a more difficult problem than the Compton continuum. "Close" here refers to a neighboring gamma-ray line whose observable peak overlaps that of the 185.7 keV line due to the finite energy resolution of the detector. A good example of this effect is found in enrichment measurements of uranium recently separated from a reprocessing plant.⁽³⁾ In this example, the peak from a strong 208 keV gamma-ray from ^{237}U (half-life 6.75 days) overlapped the 185.7 keV peak when using a NaI detector (with an energy resolution of $\text{FWHM}^* \sim 11\%$ at 208 keV). However, if a Ge(Li) detector were used (with an energy resolution of $\text{FWHM} < 1\%$ at 208 keV), both peaks would be clearly separated and resolved (see Section 3.2). Other approaches to this problem include analytical separations of two unresolved peaks (sometimes referred to as "peak stripping"). However, these techniques involve complicated analyses and are outside the scope of this guide.

Clearly, one of the most difficult problems associated with unknown interfering lines is that they may lead to erroneous enrichment results. Thus, for new types of samples, a close look at the spectrum in the vicinity of the 185.7 keV gamma ray with a multichannel analyzer is recommended to locate possible interferences before applying the standard enrichment measurement technique. To place this problem in context, it should be mentioned that most uranium samples analyzed to date have not contained materials which produce gamma-ray lines with peaks that overlap the 185.7 keV peak.

2.4.3 Ambient Backgrounds

The third source of background is different from the previous two in that it is not produced by materials in the sample. It originates from natural sources (e.g., cosmic rays) and from other radioactive materials located in the vicinity of the measuring apparatus. This last source can be particularly bothersome since it can vary in time within wide limits depending on plant operating conditions. For

*FWHM - full width of the spectrum peak at half its maximum height (see Ref. 1).

this reason, the detector itself should be adequately shielded with lead or tungsten. A good check on the efficiency of the shielding can be performed by placing radioactive sources in close proximity to the shielded detector and checking for anomalous results from standard samples.

2.5 Standard and Calibration Effects

As with any measurement technique, the importance of using good standards cannot be overemphasized. A "good" standard is one which has both a well-defined content and which is also representative of the set of samples to be measured. This second point deserves further attention since one of the more attractive features of this technique is that it is relatively independent of certain sample characteristics. An important question regarding standards is, therefore, "how many standards are required to cover a specified range of samples in order to attain a prescribed level of accuracy?" Some rough guidelines are given below to aid in answering this question.

2.5.1 Sample Composition

The measured activity is not strongly dependent on uranium density or matrix material composition for certain types of samples. Assuming that identical containers are used and that the measurement procedures are the same for all samples, the approximate ratio of the measured activities for two samples (A and B) with the same enrichment but different composition can be obtained from Eq. 2.

$$\begin{array}{l} \text{ratio of measured ac-} \\ \text{tivities} \\ \frac{\text{Sample B}}{\text{Sample A}} \end{array} = \frac{\left(1 + \sum_i \frac{\mu_i \rho_i}{\mu_u \rho_u}\right)_A}{\left(1 + \sum_i \frac{\mu_i \rho_i}{\mu_u \rho_u}\right)_B} = \frac{\left(1 + \frac{\mu_M \rho_M}{\mu_u \rho_u}\right)_A}{\left(1 + \frac{\mu_M \rho_M}{\mu_u \rho_u}\right)_B} \quad (8)$$

As an example of the use of Eq. 8, consider the differences in measured activities for pure UO_2 , U_3O_8 , and UF_6 samples:

$$\frac{\text{Activity of } \text{UO}_2}{\text{Activity of } \text{U}_3\text{O}_8} = \frac{1.015}{1.012} = 1.03$$

$$\frac{\text{Activity of } \text{UO}_2}{\text{Activity of } \text{UF}_6} = \frac{1.040}{1.012} = 1.03$$

Thus, from these results, one could expect a calibration performed using UO_2 standards to serve for U_3O_8 samples and affect the accuracy of the results by less than 1%. A bias of several percent would be introduced, however, if the UO_2 calibration were used for measurements of UF_6 samples. Whether or not this bias is acceptable depends on other conditions of the measurement. If the sample contents are known, a bias can be corrected for using Eq. 8. A special case occurs if the sample matrix is of low atomic number; then the factor (μ_M/μ_U) is approximately equal to 0.07 and only the relative densities of the uranium and matrix materials (ρ_M/ρ_U) need be known. What is especially important to note here is that an estimate of the range of applicability of any standard is available through a simple calculation.

2.5.2 Container Wall Effects

Variations in measured activity resulting from varying thicknesses of container walls is covered in Section 2.3. It is sufficient to reemphasize here that the definition of a standard and estimates of its applicability to a range of samples must include a careful consideration of the sample container. In some cases, this may turn out to be a dominating effect. An example of the importance of wall effects found in Refs. 2 and 5. Here, enrichment measurements based on the 185.7 keV gamma activity of UF_6 cylinders were found to differ by several percent from mass spectrometer data. These differences were determined to arise from variations in the thickness of the cylinder walls.

3. Equipment Specifications

Several equipment specifications important for enrichment measurements (in addition to general requirements for gamma ray spectroscopy systems) are covered in the following section.

3.1 Detector Size and Resolution

There are two important considerations in selecting a detector for the enrichment measurement application: the signal-to-background (S/B) ratio and the counting rate.

3.1.1 S/B Ratio

The S/B ratio will be poorer for lower enrichments of U^{235} . The upper curve in Fig. 4 shows the gamma spectrum of natural UO_2 taken with a 3 in. diameter x 3 in. thick sodium iodide detector. The ratio of counts in the 185 keV full energy peak (which has a FWHM of about 22 keV) to the counts in the background under it is about 0.3 for this spectrum. This ratio can be improved by using a thinner detector which is equally efficient at 185.7 keV but which produces less Compton continuum due to interactions with higher energy gamma rays. A thickness of 0.5 inches has been chosen for several enrichment meter applications. (Refs. 2 and 3.)

The lower curve in Fig. 4 shows the gamma spectrum obtained from natural UO_2 using a 50 cm³ Ge(Li) detector (approximately 4 cm thick). The signal to background for the 185 keV peak here is 1.0. This could be improved somewhat by using a thinner detector; however, the resultant loss in counting efficiency might necessitate excessively long counting times.

Figure 5 shows the calculated ideal relationship between the net peak area and its statistical uncertainty for the two (S/B) ratios obtained above. (Also shown is the case for zero background where the uncertainty in the peak count is simply the square root of the number of counts.) The NaI detector needs approximately twice as many counts as the Ge(Li)

Fig. 5. Statistical uncertainty in net counts under the 185.7 keV peak for different detectors.

detector for the same statistical uncertainty. In addition, the background subtraction is more critical and more difficult to automate for the NaI detector. These attractive features of a Ge(Li) system are offset by the relative simplicity and lower costs of a NaI system. The requirements of each specific application will determine if the improved performance attained with the Ge(Li) detector warrants the additional cost and complexity of the system.

3.1.2 Count Rate

The number of 185.7 keV gamma rays emitted from the surface of fully enriched uranium metal was calculated using Eq. 3. The result, directly proportional to the enrichment, is 2.8×10^4 gamma rays per cm^2 per second for pure (100% enrichment) ^{235}U . The gamma flux from other materials may be obtained using appropriate factors for material composition, wall effects, and enrichment as explained in Section 1. For convenience, we rewrite Eq. 3 for this special case and define the count rate in the full energy peak of a detector for bare uranium metal as (C):

$$C(\text{counts/sec}) = EA \frac{\epsilon'(\%)}{100} \left(\frac{\Omega}{4\pi}\right) 2.8 \times 10^4 \quad (9)$$

where

E = enrichment (≤ 1)

A = area of the sample viewed through the collimator (cm^2)

ϵ' = detector full energy peak efficiency (%)

$\frac{\Omega}{4\pi}$ = fractional solid angle subtended by the collimated detector.

For NaI detectors, the full energy peak efficiency is usually divided into two separate factors: (1) the total efficiency for any gamma ray interaction and (2) the photofraction (or peak-to-total ratio) which expresses the fraction of total

interactions which contribute events recorded in the photo-peak. In general, the total efficiency changes slightly with source-to-detector distance, but the peak-to-total ratio remains relatively constant. If the detector is collimated, the peak-to-total ratio will vary slightly as a function of the collimated area of the detector. Some typical values for full energy peak efficiencies are given in Table 3 for representative detectors and source-to-detector distances of 15-25 cm for 185 keV gamma rays. These values can be extrapolated to other detectors and geometries with sufficient accuracy for most count rate estimates.

Table 3
Approximate Efficiencies for 185 keV Gamma Rays*

Detector Type	Size Dia. x Length	Approximate Efficiency (%)		
		Total	Peak/Total	Full Energy Peak
NaI	3" x 3"	96	94	90
NaI	1-1/2" x 1"	88	87	76
NaI	1-1/2" x 1/2"	80	82	66
Ge (Li)	4.3 cm x 3.7 cm (~50 cc)			40

The solid angle subtended by the detector divided by 4π (defined here as the fractional solid angle) for a circular detector is given in Fig. 6 as a function of the ratio of the radius of the detector to the source-to-detector distance. The upper curve applies to a point source and uncollimated detector and the lower curve applies to a cylindrical collimator with a finite source covering one end and the detector covering the other end.

*Scintillation Phosphors (second edition, 1969), E. C. Stewart, et al, Harshaw Chemical Company; F. C. Young, et al., Nuc. Instr. and Methods, 92, pp. 71-75 (1971).

Fig. 6. Fractional solid angles subtended by a circular area detector for point source (uncollimated) and a finite source (cylindrical collimator) as a function of the source to detector distance (D) and the radius of the exposed area of the detector (R). (From R. P. Gardner and k. Verghese, Nuc. Instr. and Methods, 93, No. 1, pp. 163-167 (1971).)

As an example of the use of Eq. 9, a 1-1/2 in. diameter by 1/2 in. thick NaI detector placed directly against a uranium metal plate enriched to 93% ^{235}U , will result in a count rate for the 185.7 keV peak of about 1.0×10^5 counts per second (this rate, incidentally would introduce severe dead time losses in the measured counting rate). Using the same Eq. 9, if one were to collimate the same NaI detector with a 0.5 cm thick, 1 cm diameter circular tungsten collimator, the count rate for the 185.7 keV peak would be about 4.2×10^2 counts per second--an acceptable rate for most systems.*

Calculation of the detector count rates for purposes of making dead time estimates requires that one calculate the total count rate. For the case of highly enriched uranium (above), the 185.7 keV full energy peak counts are responsible for about 80% or more of the total count rate. The total count rate is therefore approximately equal to the 185.7 keV peak count rate. At the other end of the enrichment scale, for naturally enriched uranium (0.7%) the 185.7 keV peak counts are responsible for only about 5% to 10% of the total count rate. Total count rate estimates for low enriched material must therefore take into account this relatively important background from ^{238}U gamma and x-rays. If other radioactive materials are present within the sample, their contributions to the total count rate must also be considered.

In general, the count rate should be limited so that losses due to dead time in the counting system can be accounted for within the desired measurement uncertainty. These losses can be taken into account in several different ways: (1) If they are known to be small compared to the overall uncertainty desired, then they can be neglected. (2) Standards can be used to determine the non-linear calibration curve of counts (including dead time losses) vs enrichment. (3) Instruments can be used which electronically correct for dead time and measurements are then made for known

* Measured count rates can differ from those calculated with these simple models since all the physical effects such as collimator scattering, details of background subtraction, penumbra, etc., are not included. Estimates derived using Eq. 9 should agree with measured results to within about a factor of 2.

"live time"* interval. The latter method is used in most multichannel analyzer systems. For a simple single channel analyzer arrangement with a typical pulse pair resolution of 2 μ sec, dead time losses will be about 2% at a total count rate of 10,000 counts per second. This count rate should be considered an upper limit unless some type of dead time correction is used.

Dead time effects can be eliminated in certain cases. For example, samples with thin container walls can emit large numbers of low energy x-rays (≤ 100 keV) which contribute to high count rate problems, but provide no useful information. These x-rays can be strongly absorbed without significantly attenuating the 185.7 keV gamma ray flux incident on the detector by covering the NaI crystal with a cadmium filter, 0.076 to 0.15 cm (0.03 to 0.06 in.) thick.

Pulse pile-up, a problem which also results in a loss of counts in the full energy peak, can be important for Ge(Li) systems. Pulse pile-up occurs when one detected gamma ray pulse (A) overlaps the "tail" of an adjoining pulse (the frequency of occurrence of this event obviously rises with increasing count rate). This causes the pulse height of event (A) to be greater than its equivalent gamma ray energy. For high resolution Ge(Li) systems, this can result in the event being recorded outside the full energy peak. This loss of counts in the full energy peak can best be corrected for by introducing a pulser peak with a known rate into the spectrum and measuring the loss of counts in the pulser peak due to this effect.** Reference 1 contains a more complete discussion of losses in counting rates due to pulse pile-up.

3.2 ^{237}U Interference

The typical NaI detector resolution of about 22 keV FWHM for 185.7 keV gamma rays has usually proved to be adequate for

- * live-time - that portion of the measurement period during which the instrument can record detected events. Dead time refers to that portion of the measurement period during which the instrument is busy processing data already received and cannot accept new data. In order to compare different data for which dead times are appreciable, one must compare counts measured for equal live time periods. (actual measurement period) - (dead time) = live time
- ** The reduction in a pulser peak due to pulse pile-up will not be exactly the same as the reduction in the 185.7 keV peak unless the amplifier pulse shapes are identical.

enrichment meter applications within the accuracy and signal/background considerations discussed above. One application for which NaI is inadequate is the measurement of reprocessed uranium containing significant amounts of ^{237}U . ^{237}U , a decay product of ^{241}Pu , emits several gamma rays, the most intense of which has an energy of 208 keV. Figure 7 shows the spectra of natural UO_2 with different ^{237}U content measured with a 3 in. x 3 in. NaI detector. The bottom spectrum results from uranium containing no ^{237}U with the ^{235}U peak superimposed on the Compton continuum background. The center spectrum shows the effect of adding about 0.035 Ci of ^{237}U activity per gram uranium to the sample and the 208 keV peak count rate is about equal to the 185.7 keV peak from ^{235}U . The upper spectrum shows the effect of about 0.2 Ci of ^{237}U per gram uranium activity and clearly illustrates the interference with the 185.7 keV peak.

The ^{237}U activity present in reprocessed uranium will depend on the amount of ^{241}Pu present before reprocessing and the time elapsed since separation (^{237}U has a half-life of 6.75 days). Figure 8 shows the 208 keV activity from ^{237}U as a function of the time after separation. This calculation was made for high burn-up (30,000 MWD/MTU) LWR fuel which contained 15 mg Pu (1.8 mg ^{241}Pu) per gram uranium. The 208 keV activity is clearly a problem for this material, reaching the same intensity as the 185.7 keV activity about 43 days after separation from Pu, assuming 1% of the remaining uranium is ^{235}U .

Figure 9 shows the gamma ray spectra from this material using a 50 cm³ Ge(Li) detector. Due to the improved resolution of the Ge(Li) system, there is no interference with the 185.7 keV peak at the ^{237}U levels shown here. For freshly reprocessed material with a 208 keV activity 100 times that of the 185.7 keV peak from ^{235}U , there might be some interference depending on the shape of the 208 keV peak. The degree of interference would depend on the specific detector, count rate, electronics, etc., and must be determined experimentally.

Fig. 7. NaI gamma ray spectra of natural uranium oxide with ^{237}U peaks simulated.

- A) No ^{237}U
- B) $0.035 \mu\text{Ci/gU } ^{237}\text{U}$ activity
- C) $0.17 \mu\text{Ci/gU } ^{237}\text{U}$ activity

Fig. 8. 208 keV activity from ^{237}U ($T_{1/2} = 6.75$ days) following Pu separation.

Fig. 9. Gamma ray spectra (Ge(Li)) of natural UO_2 with simulated ^{237}U contents.

- A) no ^{237}U
- B) 0.035 $\mu\text{Ci/gU}$ ^{237}U activity
- C) 0.35 $\mu\text{Ci/gU}$ ^{237}U activity

3.3 Instrument Stability

Most modern counting electronics operate quite stably for extended periods of time. Photomultiplier tubes are, however, sensitive to changes in temperature, count rate, magnetic field, etc., and can produce important inaccuracies due to gain shifts. Some provision should be made for gain checks and/or gain stabilization for enrichment measurement applications.

In the case of multiple single channel analyzer systems, a variety of gain stabilizers are available that automatically adjust the system gain to keep a reference peak centered between two preset energy limits. The 185.7 keV peak itself could be used as a reference; however, difficulties arise when the peak count rate is low and the peak is located on a sloping background. A stabilizing technique that has often been used with a NaI detector and multiple single channel analyzers is the NaI crystal with a built-in reference peak. These are constructed by doping a portion of the crystal with an alpha-particle source material (e.g., ^{241}Am). The alpha particles provide adequate count rates in a peak located at an equivalent gamma ray of a few MeV, creating virtually no interference with lower energy peaks. This peak is located on a low continuum background and an optimal count rate is selected by choosing an appropriate alpha-particle source strength.

It should be noted, however, that the gamma equivalent energy of the alpha pulse (approximately 3.5 MeV) is known to shift with temperature. In some units, this shift has been electronically compensated for so that stabilization of the apparatus, based on the location of the alpha peak, is still valid. However, for applications during which the apparatus is subjected to a significant temperature shift ($> 10^{\circ}\text{C}$), it should be recalibrated at the new temperature after sufficient time has elapsed for the unit to arrive at thermal equilibrium with its surroundings.

A second technique for producing a useful reference peak, whose position is not subject to temperature drifts relative to the gamma ray peak, is to use an external gamma ray source. ^{241}Am produces a 60 keV gamma ray which is sufficiently lower

in energy so that its peak does not overlap that of the 185.7 keV line. The source is placed between the NaI crystal and a cadmium filter, so that any low energy x-ray background from the sample does not interfere with the 60 keV reference peak.

In the case of "saturated" semiconductor detectors, the signal is directly related to the energy of the absorbed gamma ray and a stable pulse generator attached to the pre-amplifier may be used to provide a reference for gain stabilization.

A variety of commercial gain stabilizing units are available for use with NaI or Ge(Li) detectors and several classes of amplifier-analyzer systems. The point to bear in mind is that detector and/or amplifier gains are likely to drift during use and that methods are available to reduce such drifts.

4. Data Storage and Analysis

It was assumed in the derivation of the basic equation describing this measurement (Eq. 3) that the enrichment of the sample was proportional to the net number of counts contained in the full energy peak of the 185.7 keV line. All data analysis techniques are designed, therefore, to determine the net area under this peak in the presence of backgrounds. (Backgrounds are described in Section 2.4.) This section describes two different data storage and related analysis techniques which include background subtraction.

4.1 Multiple Single Channel Analyzer

In recent experience, most systems that have used the multiple single-channel analyzer approach for data storage and analysis have employed a NaI detector. For this reason, the following discussion specifically refers to a NaI system; however the general remarks (but not the detailed specifications) could apply equally as well to a Ge(Li) system.

A typical spectrum from a NaI detector is shown in Fig. 10. The 185.7 keV full energy peak superimposed on a smooth continuous background is clearly identified. The net counts in the 185.7 keV peak are determined by setting two single-channel analyzers to pass only those detector pulses falling within two different energy intervals (1 and 2) and two separate scalars (1 and 2 respectively). The pulses in energy interval 1 contain the net counts in the 185.7 keV peak plus the counts in the continuous background under the peak. The counts in energy interval 2 contain only those counts in the continuous background present in this interval. Typical NaI settings for the energy intervals 1 and 2 are 160-210 keV and 220-270 keV respectively (Refs. 2 and 4).

The enrichment (E) of an unknown sample is determined from the expression,

$$E = aC_1 - bC_2 \quad (10)$$

$$\Delta E/E (\%) = \frac{\sqrt{a^2 C_1 + b^2 C_2}}{aC_1 - bC_2} \times 100 \quad (11)$$

Fig. 10. Multiple single channel analyzer approach for determining the net counts in the 185.7 keV peak.

$C_{1,2}$ = counts stored in scalars 1, 2

a, b = constants experimentally determined from the measurement of two standards of known and different enrichments (see Appendix 3 for a discussion of calibration techniques)

$\Delta E/E$ = statistical uncertainty in the enrichment*

In order to relate this expression to the discussion above, it is instructive to write Eq. 10 as follows,

$$E = a \left(C_1 - \frac{b}{a} C_2 \right) \quad . \quad (12)$$

This expression can be interpreted as,

E = calibration constant (a) x net counts in 185.7 keV peak

The constant (b/a) is therefore the ratio of the contribution of the continuum background in interval 1 to the contribution of the continuum in window 2. The constant (a) is a calibration constant which takes into account the detector parameters, container wall effects, physical constants, and material composition.

Two potential problems can be identified from the discussion above:

- It is assumed that the ratio of the backgrounds (b/a) in energy intervals 1 and 2 are constant over the range of samples of interest. Changes in the shape of the background produced by unexpected sources of radioactivity within a sample or other effects can change this ratio and produce erroneous results (see Section 2.4.1).

*The statistical uncertainty represents the minimum uncertainty in the measurement that can be attained (due to the finite number of counts). The total uncertainty in the measurement can be significantly larger than this and should always be determined empirically.

- Instabilities of the system electronics can result in variations of limits of the two energy intervals and thus produce changes in the ratio (b/a) . It is important, therefore, to periodically check the calibration of the instrument with a standard sample.

Several variations of multiple single-channel analyzer systems have been used in the past. Some versions use an "up-down scalar" which automatically subtracts the continuum background and displays the result $C = C_1 - C_2$. Another version uses an up-down scalar plus an additional control to adjust the constants (a) and (b) so that the enrichment $E = a \times (C_1 - b/a C_2)$ is displayed directly. These systems do not differ in principle from the "double scalar" system described above in which both C_1 and C_2 are displayed.

4.2 Multichannel Analyzers

Most multichannel analyzers have a display so that the shape of the spectrum can be visually checked. Spectra of standards and unknowns may be compared to determine whether they are similar or unlike in some respect which might affect the calibration (as discussed in Section 2). Gain drifts and other malfunctions are readily determined.

A variety of schemes have been employed to determine the net count in gamma ray peaks using either NaI or Ge(Li) detectors and a multichannel analyzer ranging from relatively simple schemes and hand calculations to elaborate programs for on-line or off-line computers. Sophistication is probably not justified for uranium enrichment measurements. System gains should be adjusted so that 10 or more channels contain the 185.7 keV peak and a comparable number of channels should be used to define the continuum which is to be subtracted to obtain the net counts of interest.

Fig. 11 shows one method which may be used to obtain the net count with a NaI detector. The actual net count is the shaded area shown in the top figure. An approximation to this is obtained as shown in the lower figure by

Fig. 11. Net counts under the 185.7 keV peak. (A) actual. (B) as determined by Eq. 13.

linearly extending the sides of the peak until they intersect a straight line (backward) extrapolation of the baseline continuum. This determines boundary channels m and n for determining the gross count of the peak. T channels above n and below m are summed to give the average counts for a channel $\bar{c}_m$ below m and a channel $\bar{c}_n$ above n . The rectangular area $[(\bar{c}_m + \bar{c}_n)/2] \times (m - n + 1)$ is used to represent the continuum under the peak. The enrichment is given to a close approximation by

$$E = K \left[\sum_{i=m}^n c_i - \left(\frac{\bar{c}_m + \bar{c}_n}{2} \right) (m - n + 1) \right]$$

= calibration constant x net counts in 185.7 keV peak

(13)

The statistical precision is

$$\frac{\Delta E}{E} (\%) = \frac{\sqrt{\sum c_i + \frac{(m - n + 1)^2}{4T} (\bar{c}_m + \bar{c}_n)}}{\sum c_i - \left(\frac{\bar{c}_m + \bar{c}_n}{2} \right) (m - n + 1)} \times 100$$
(14)

where

c_i = total counts in channel i

K = constant determined by calibration

$\Delta E/E$ = statistical uncertainty in the enrichment (see note under Eq. 11)

The actual precision will be somewhat poorer than that given by Equation 14 since factors other than counting statistics are involved. Precision should be determined empirically by making repeated runs on standards and samples. Ref. 7 describes a system which is somewhat similar to this example and specifies the procedures for calibration and error analysis.

Other techniques can include peak fitting and non-linear fits to the continuum; these involve much more elaborate computing capabilities and are probably not necessary for most enrichment applications.

5. Applications

The discussions in the preceding sections have been general in the sense that they have not dealt with specific samples or circumstances peculiar to a given measurement problem. This section presents reported results for specific cases. The intention is to illustrate the wide diversity of applications for which this technique can be useful; it is not meant to represent a complete record of all the work performed to date using variations of the basic technique. These results also illustrate several of the problems which were discussed in more general terms in Section 2.

The representative data are given below in Table 4.

The results presented are self-explanatory. It is worthwhile to point out, however, that the accuracy of the technique can vary within wide limits depending on the sample description and the measurement system. Accuracies of $\pm 10\%$ (relative) may be adequate for quick checks of sealed containers to verify that they hold high enriched material, while accuracies on the order of 1 or 2% (relative) may be required when fuel pellets are randomly sampled to quickly confirm an enrichment previously assigned on the basis of mass spectrometer measurements. The associated measurement system can range from a simple, portable, hand-held NaI detector, to a permanent on-line computer controlled installation. It would be impossible to cover all conceivable applications of this enrichment technique. Rather, each new measurement problem should be approached from the standpoint that an optimal measurement system can be designed and the performance of that system can be accurately defined. The purpose of the guideline is to assist in performing that task.

Examples of Results from the Application of the 185.7 keV Gamma Ray
Enrichment Measurement to Fuel Cycle Samples

DESCRIPTION	SAMPLE CONTAINER	ENRICHMENT RANGE	PERFORMANCE	COMMENTS	DETECTOR	REFERENCE
Incinerator oxides ≥ 80 w/o U	Cans; 15 in. H, 6 in. OD	1 to 5%	5 minute count period, $\sim 1\%$ statistical uncertainty, estimated relative accuracy $\pm 5\%$.	Errors caused by stratification of enrichment in cans are discussed.	Ge(Li)	5
Fuel Rods	0.4 to 2.8 in. D; 2 to 10 ft L.	3 to 10%	5 min count period. Average difference from assigned values 4.5%.	Uncertainties due to variations in cladding thickness unknown.	NaI	5
UO ₂ Pellets	2 gallon cardboard drums, 10 in. O.D.	2 to 4%	5 min count period. 1% statistical uncertainty.	Accuracy limited by mixtures of pellets with different enrichments.	Ge(Li)	5
UF ₆	Cylinders; 5 in. D, 30 in. H (1/4 in. Monel) 48 in. D, 10 ft. H (5/8 in. steel)	3 to 98%	Results within $\pm 10\%$ (relative) of assigned values.	Useful for spot checks on cylinder enrichment.	NaI (portable)	2
UF ₆	Product cylinders; 5 in. D, 30 in. H	20 to 98%	Estimated uncertainty in a single measurement $< 10\%$ (relative).	Uncertainties caused by variations in cylinder wall thickness are discussed.	Ge(Li)	6
UF ₆	Cell inlet pipes	5 to 80%	Results within several percent (relative) of assigned values.	Further study required to determine if material deposit collecting on pipe walls are present. Example of samples which are not of semi-infinite thickness. Pipe diameters and gas pressure required to calculate enrichment from measured count rate.	NaI (portable)	2
UF ₆ (liquid)	Loadout pipe		20 counts/sec percent ²³⁵ U, 1% statistical uncertainty. Relative difference from mass spec measurements over 3 week period; .006 std. deviation .027 mean deviation	Results describe performance of prototype. Permanent system (Ref. 6) will have count rate 5 to 10 times higher.	NaI	3, 6
UF ₆	2-1/2 ton UF ₆ cylinders	1.3 to 3.1%	300 sec count period. Standard deviation from assigned values was 5.3% (80 cylinders). However 17 cylinders were found to have differences in excess of 16%.	²³⁴ Th may be plating out on cylinder surfaces, and thus change the shape of the background continuum for these particular cylinders.	NaI	8
UO ₂ product and feed material	5 gal. bucket	0.2 to 5%	400 sec. count period. Average difference from assigned values $\sim 1.7\%$.	Part of hybrid system which includes neutron counters to determine ²³⁵ U content.	NaI	4
Individual UO ₂ Pellets	Pellets 1 to 4 cm D; 1 to 2 cm H (7 to 100 gm)	1 to 16%	6 min count period. Average relative deviation from mass spec measurements was about 2% for a 3% enrichment		NaI	7

Appendix 1

Gamma Ray Attenuation Coefficients-Definitions

The attenuation of an initial gamma ray flux I_0 (gammas/cm² . sec) of a given energy (E_γ) in passing a distance x (cm) through a material of density ρ (g/cm³) is given by

$$\frac{I(x)}{I_0} = \exp(-\mu\rho x) \quad (A1-1)$$

where

$I(x)$ = gamma ray flux at position x

μ = gamma ray attenuation coefficient (cm²/g)

If the material is made up of a number of elements ($i = 1, 2, 3, \dots$), then

$$\mu\rho = \sum_i \mu_i \rho_i \quad (A1-2)$$

where μ_i , ρ_i are the attenuation coefficients and densities respectively of all the individual elements making up the composite material. In several discussions included in this guide, it is convenient to split the attenuation coefficient for sample materials into two parts: (1) uranium and (2) all other materials which make up the sample matrix. Analytically,

$$\begin{aligned} \mu\rho &= \mu_u \rho_u + \sum_j \mu_j \rho_j \\ &= \mu_u \rho_u + \mu_M \rho_M \end{aligned} \quad (A1-3)$$

where

μ_u, ρ_u = mass attenuation coefficient (cm^2/g),
density (g/cm^3) for uranium

μ_j, ρ_j = mass attenuation coefficient, density for
all other components of the matrix ($i = 1, 2, 3, \dots$)

μ_M, ρ_M = mass attenuation coefficient, density for the
composite matrix ($\mu_M \rho_M = \sum_j \mu_j \rho_j$)

Mass attenuation data are given for each element and for three types of attenuation: elastic scattering, inelastic scattering and true absorption. In these calculations the elastic scattering factor should be ignored because as many gamma rays should be scattered into the region viewed by the collimated detector as are scattered out. Those totally absorbed and those that lose some energy for example by the Compton process are lost from the 185.7 keV peak.

Some insight into the physical significance of the coefficient, $\mu\rho$, can be obtained by calculating the value of the mean distance traveled (λ) before scattering or absorption for a gamma ray of energy E_γ incident on a material of density ρ with attenuation coefficient μ . The result is simply the inverse of the absorption coefficient.

$$\lambda(\text{cm}) = \frac{1}{\mu\rho} = \frac{1}{\mu}, \quad . \quad (\text{A1-5})$$

Appendix 2

Analytical Expressions for Estimating the Effect of Varying Sample Thickness on Measured Activity

The percentage difference introduced in the measured count rate ($\Delta C/C$) by a difference in thickness Δx (cm) of a sample of thickness x (cm) is given by

$$\frac{\Delta C}{C} (\%) = (\mu' \Delta x) f(\mu' x) \quad (A2-1)$$

where

$$\mu' = \mu_u \rho_u + \sum_i \mu_i \rho_i$$

$$f(\mu' x) = \frac{e^{-\mu' x}}{1 - e^{-\mu' x}} \times 100 \quad (A2-2)$$

This expression is given to estimate the measurement sensitivity to variations in sample depth in the event that measurements on thin samples are required. The function $f(\mu' x)$ is plotted in Fig. A2-1 for convenience.

Fig. A2-1. The function $f(\mu'x)$ from Eq. A2-2.

Appendix 3

Calibration Procedures for Enrichment Measurements with Multiple Single- Channel Systems

The following general procedure is recommended for calibrating multiple single-channel systems. Details associated with specific equipment are omitted. Definitions of terms used are given in Section 4.1, Fig. 10. The following terms are also used in this discussion.

- Threshold 1: lower energy limit of energy window 1
- Threshold 2: lower energy limit of energy window 2
- Window width 1: width of energy window 1 = upper energy limit of energy window 1 minus Threshold 1
- Window width 2: width of energy window 2 = upper energy limit of energy window 2 minus Threshold 2

As an example of this terminology, consider a typical energy interval (1), of 160 to 210 keV for single channel analyzer (1) which passes counts to scalar (1). Threshold (1) is therefore 160 keV and window width (1) is $210 - 160 = 50$ keV.

Calibration Steps

- Step 1 Set window width 1 slightly wider than the detector resolution at 185 keV. Set threshold 1 so as to maximize the count rate from the 185.7 keV line.
- Step 2 Set window width 2 approximately equal to window width 1 (for systems with up-down scalars, window width 2 is set approximately equal to 1.3 to 1.4 times window width 1). Threshold 2 should be set about 1.4 times higher than threshold 1.

Step 3 Select two standards (1,2) in accordance with the criteria specified in Section 2.5. The enrichments of the standards should represent the upper and lower extremes of the enrichment range of interest. Record C_{11} , C_{21} * counts in scalars 1 and 2 respectively for standard 1 and C_{12} and C_{22} counts in scalars 1 and 2 for standard 2.

Step 4 The known enrichments (E_1 , E_2) of standards 1 and 2 are then given by

$$E_1 = a C_{11} - b C_{21} \quad (A3-1)$$

$$E_2 = a C_{12} - b C_{22} \quad (A3-2)$$

where a and b are constants. Equations A3-1 and A3-2 can be solved for a and b with the following result:

$$- \frac{E_1 C_{12} - E_2 C_{11}}{C_{21} C_{12} - C_{22} C_{11}} = b \quad (A3-3)$$

$$\frac{E_1 C_{22} - E_2 C_{21}}{C_{11} C_{22} - C_{12} C_{21}} = a \quad (A3-4)$$

Step 5 Enrichments of unknown samples can then be determined from the relation:

$$E = a C_1 - b C_2$$

where a and b are given above.

*Note: C_{21} refers to scalar 2 and sample 1. Other C_{ij} are defined in a similar manner.

Note 1 If up-down scalars are used, window width 2 should be adjusted so that $b/a = 1$. The most practical method for accomplishing this is to use a depleted uranium standard (enrichment ~ 0) and adjust window width 2 such that $C_1 = C_2$ for the depleted standard. This method is not exact since depleted uranium exhibits some 185.7 keV activity, and introduces a bias into the calibration. This may be relatively unimportant for high enrichment measurements but could be significant for assays of low enrichment samples.

Note 2 If excessive count rates are present the relation between count rate and enrichment will not be linear, and the above technique is not applicable. In this case, one can derive a non-linear calibration using three or more standards which cover the range, or limit the count rate with collimators or filters. If the enrichment range of interest is limited to about 5 or 10%, the count rate should be limited so that the calibration is at least linear over this range of interest.

Note 3 Calibration errors. The calibration constants (a and b) have a minimum uncertainty defined by the statistical uncertainty of the counts (C_{11} , C_{21} , C_{12} , C_{22}) recorded during the calibration run(s) and the uncertainty in the known enrichments of the standards (E_1 , E_2). Calibration runs should therefore collect a minimum of 10^5 counts in scalar 1 in order to minimize the statistical uncertainty in the calibration. It is important to remember, however, that the uncertainty in the measurement of an unknown depends not only on the calibration errors, but also on the instrument stability and applicability of the standards to the unknown. These additional uncertainties often dominate the measurement and their magnitude can only be determined empirically.

6. References

1. Kull, L. A., "An Introduction to Ge(li) and NaI Gamma Ray Detectors for Safeguards Applications," ANL-ACEA 103, 1973.
2. Reilly, T.D. and Walton, R. B., "Nuclear Safeguards Research and Development," LA-4705 MS, 1971 (p. 16).
3. Reilly, T.D. and Walton, R. B., "Nuclear Analysis Research and Development," LA-5091 PR, 1972 (p. 19).
4. Reilly, T. D., Walton, R. B., Martin, E. R. and Parker, J. L., "Nuclear Analysis Research and Development," LA-5197 PR, 1972 (p. 11).
5. Parker, J. L., "Nuclear Safeguards Research and Development," LA-4605, 1970 (p. 36).
6. Reilly, T. D., Parker, J. L., Evans, A. E., Walton, R. B., "Nuclear Safeguards Research and Development," LA-4794, 1971 (p. 16).
7. Gunderson, G., Cohen, I., Zucker, M., Proceedings: 13th Annual Meeting, Institute of Nuclear Materials Management, (1972) p. 221.
8. Walton, R. B., Reilly, T. D., Parker, J. L. and Menzel, J. H., "Measurements of UF₆ Cylinders with Portable Instruments," LA-UR-73-880, 1973. Proceedings: 14th Annual Meeting, Institute of Nuclear Materials Management (1973) p. 320.
9. Handbook of Chemistry and Physics, C. D. Hodgman (Ed.), 41st Edition, Chemical Rubber Publ. Co., 1960.
10. Gamma Ray Attenuation Constants taken from E. F. Plechaty, J. R. Terral, "Photon Cross Sections 1 keV to 100 MeV," TID-4500, 1968.
11. "Photon Cross Sections, Attenuation Coefficients and Energy Absorption Coefficients from 10 keV to 100 GeV," N-SRDS-NBS 29 (1969).

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