April 1974

U.S. ATOMIC ENERGY COMMISSION

REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

REGULATORY GUIDE 5.21

NONDESTRUCTIVE URANIUM-235 ENRICHMENT ASSAY BY GAMMA-RAY SPECTROMETRY

A. INTRODUCTION

Section 70.51, "Material Balance, Inventory, and Records Requirements," of 10 CFR Part 70, "Special Nuclear Material," requires, in part, that licensees authorized to possess at any one time more than one effective kilogram of special nuclear material (SNM) determine the material unaccounted for (MUF) and its associated limit of error (LEMUF) for each element and the fissile isotope for uranium contained in material in process. Such a determination is to be based on measurements of the quantity of the element and of the fissile isotope for uranium.

The majority of measurement techniques used in SNM accountability are specific to either the element or the isotope but not to both. A combination of techniques is therefore required to determine the MUF and LEMUF by element and by fissile isotope for uranium. Passive gamma-ray spectrometry is a nondestructive method for measuring the enrichment, or relative concentration, of the fissile isotope U-235 in uranium. As such, this technique is used in conjunction with an assay for the element uranium in order to determine the amount of U-235.

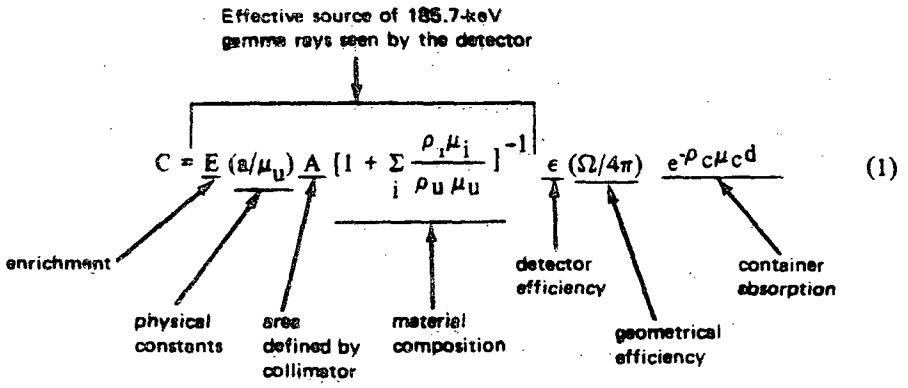
This guide details conditions for an acceptable U-235 enrichment measurement using gamma-ray spectrometry, and prescribes procedures for operation, calibration, error analysis, and measurement control.

B. DISCUSSION

The alpha decay of U-235 to Th-231 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 U-235). The relatively low energy and consequent low penetrating power of these gamma rays implies that most of those emitted within the interior of the material are absorbed within the material itself. These thick [1] materials therefore exhibit a 185.7-keV gamma ray activity which approximates the activity characteristic of an infinite medium: i.e., the activity does not depend on the size or dimensions of the material. Under these conditions, the 185.7-keV activity is directly proportional to the U-235 enrichment. A measurement of this 185.7-keV activity with a suitable detector forms the basis for an enrichment measurement technique.

The thickness of the material with respect to the mean free path of the 185.7-keV gamma ray is the primary characteristic which determines the applicability of passive gamma-ray spectrometry for the measurement of isotope enrichment. The enrichment technique is applicable only if the material is thick. However, in addition to the thickness of the material, other conditions must be satisfied before the gamma-ray enrichment technique can be accurately applied. An approximate analytical expression for the detected 185.7-keV activity is given below. This expression has been separated into several individual terms in order to aid in identifying those parameters which may interfere with the measurement. Although approximate, this relationship can be used to estimate the magnitude of interfering effects in order to establish limits on the range of applicability and to determine the associated uncertainties introduced into the measurement. This relationship is:

[1] "Thick" and "thin" are used throughout this guide to refer to distances in relation to the mean free path of the 185.7 keV gamma ray in the material under consideration. The mean free path is the 1/e-folding distance of the gamma-ray flux or, in other terms, the average distance a gamma ray traverses before interacting.



where

C = detected 185.7-keV activity
E = enrichment of the uranium (≤ 1)
ρ_u, ρ_i, ρ_c = density of the uranium (u), matrix material (i), and container wall (c), respectively, in (g/cm^3)
μ_u, μ_i, μ_c = mass attenuation coefficient for 185.7-keV gamma rays in uranium (u), matrix material (i), and container wall (c) in units of (cm^2/g)
a = specific 185.7-keV gamma ray activity of U-235
= 4.3×10^4 gamma rays/sec-g
ϵ = net absolute detector full energy peak efficiency for detecting 185.7-keV gamma rays (≤ 1)
Ω = solid angle subtended by the detector ($\Omega \leq 2\pi$)
A = cross-sectional area of material defined by the detector collimator
d = container wall thickness

A derivation of this expression, as well as other necessary background information relevant to this guide, may be found in the literature. [2] As evident in Eq. 1, the activity (C) is proportional to the enrichment (E) but is affected by several other characteristics as well.

Material Thickness Effects

In order for Eq. 1 to be applicable, it is necessary that the material be sufficiently thick to produce strong attenuation of 185.7-keV gamma rays. To determine whether this criterion is met, it is useful to compare the actual thickness of the material with a characteristic length x_o, where x_o is defined as that thickness of material which produces 99.5% of the measured 185.7-keV activity, i.e.,

$$x_o = -\lambda \ln(.005) = 5.29\,\lambda \qquad (2)$$

where

$$1/\lambda = \mu_u\rho_u + \sum_i \mu_i\rho_i \qquad (3)$$

Calculated values of x_o, the critical distance, for several common materials are given in Table 1.

TABLE 1[3]

Material	Density (g/cm^3)	Critical Distance x_o (cm)	Material Composition Term $1 + \sum_i \frac{\rho_i \mu_i}{\rho_u \mu_u}$
U (metal)	18.7	0.20	1.000
UF_6	4.7	1.08	1.040
UO_2	10.9	0.37	1.012
U_3O_8	7.3	0.56	1.015
Uranyl Nitrate	2.8	2.30	1.095

[2] L. A. Kull, "Guidelines for Gamma-Ray Spectroscopy Measurements of U-235 Enrichment," BNL-50414, July 1973.

[3] Values of the mass attenuation coefficient, μ, may be found in J. H. Hubbell, "Photon Cross Sections, Attenuation Coefficients, and Energy Absorption Coefficents From 10 keV to 100 GeV," NSRDS-NBS 29, 1969.

Note: Other nondestructive techniques are capable of detecting SNM distributed within a container. The enrichment technique, however, is inherently a surface measurement. Therefore, the "sample" observed i.e., the surface, must be representative of all the material in the container. In this respect the enrichment measurement is more analogous to chemical analysis than other NDA techniques.

Material Composition Effects

If the gamma-ray measurement is to be dependent only on the enrichment, the term related to the composition of the matrix should be approximately equal to one, i.e.,

$$1 + \sum_i \frac{\rho_i \mu_i}{\rho_u \mu_u} \simeq 1 \qquad (4)$$

Calculated values of this quantity for common materials are given in Table 1. The deviation of the numbers in Table 1 from unity indicate that a bias can be introduced by ignoring the difference in material composition.

Inhomogeneities in matrix material composition, uranium density, and uranium enrichment within the measured volume of the material (as characterized by the depth x_o and the collimated area A) can produce changes in the measured 185.7-keV activity and affect the accuracy of an enrichment calculated on the basis of that activity. There is a small to negligible effect on the measurement accuracy due to variations in the content of low-atomic-number ($Z<30$) matrix materials. Care should be exercised, however, in applying this technique to materials having high-atomic-number matrices ($Z>50$) or materials having uranium concentrations less than approximately 75%. Inhomogeneities in uranium density will also produce small to negligible effects on the accuracy if the matrix is of low-atomic-number elements. ***Significant inaccuracies can arise, however, when the uranium enrichment itself can be expected to vary throughout the sample.***

The above conclusions about the effects of inhomogeneities are based on the assumption that the thickness of the material exceeds the critical distance, x_o, and that the inhomogeneities exist within this depth. In the case of extremely inhomogeneous materials such as scrap, the condition of sufficient depth may not always be fulfilled, or inhomogeneities may exist beyond the depth x_o; i.e., the "sample" is not representative. Therefore, this technique is not applicable to such inhomogeneous materials.

Container Wall Effects

Variations in the thickness of the container walls can significantly affect the activity measured by the detector. The fractional change in the measured activity $\Delta C/C$ due to a small change Δd in the container wall thickness can be expressed as follows:

$$\frac{\Delta C}{C} = -\mu_c \rho_c \Delta d \qquad (5)$$

Calculated values of $\Delta C/C$, corresponding to a change in container thickness Δd of 0.0025 cm, for common container materials, are given in Table 2.

TABLE 2

Material	Density (g/cm^3)	$\frac{\Delta C}{C}$
Steel	7.8	− .003
Aluminum	2.7	− .0009
Polyethylene	0.95	− .0004

Therefore, the container wall thickness should be known, e.g., by measuring an adequate number of the containers before loading. In some cases an unknown container wall thickness can be measured using an ultrasonic technique and a simple correction applied to the data to account for attenuation of the 185.7-keV gamma rays (see eq. 5). Commercial equipment is available to measure wall thicknesses ranging from about 0.025 to 5.0 cm to relative accuracies of approximately 1.0% to 0.1%, respectively.

Area and Geometrical Efficiency

The area of the material viewed by the detector and the geometrical efficiency are variables which may be adjusted, within limits, to optimize a system. It is important to be aware that once these variables are fixed, changes in these parameters will affect the results of the measurement.

It is also important to note that the placement of the material within the container will affect the detected activity. The material should fill the volume of the container to a certain depth, leaving no void spaces between the material and the container wall.

Net Detector Efficiency

Thallium-activated sodium iodide, NaI(Tl), scintillation detectors and lithium-drifted germanium, Ge(Li), solid-state detectors have been used to perform these measurements. The detection systems are generally conventional gamma-ray spectrometry systems presently commercially available in modular or single-unit construction.

The following factors influence detector selection and the control required for accurate results.

1. **Background**

a. **Compton Background.** This background is predominately produced by the 765-keV and 1001-keV gamma rays of Pa-234m, a daughter of U-238. Since, in most cases, the Compton background behaves smoothly in the vicinity of the 185.7-keV peak, it can be readily subtracted, leaving only the net counts in the 185.7-keV full-energy peak.

b. **Overlapping Peaks.** The observable peak from certain gamma rays may overlap that of the 185.7-keV peak due to the finite energy resolution of the detector; i.e., the difference in energies may be less than twice the FWHM.[4] This problem is common in enrichment measurements of recently separated uranium from a reprocessing plant. The peak from a strong 208-keV gamma ray from U-237 (half-life of 6.75 days) can overlap the 185.7-keV peak when an NaI detector is used. Analytical separation of the two unresolved peaks, i.e., peak stripping, may be applied. An alternative solution is to use a Ge(Li) detector so that both peaks are clearly resolved.

The U-237 activity present in reprocessed uranium will depend on the amount of Pu-241 present before reprocessing and also on the time elapsed since separation.

c. **Ambient Background.** The third source of background originates from natural sources and from other uranium-bearing materials located in the vicinity of the measuring apparatus. This last source can be particularly bothersome since it can vary with time within wide limits depending on plant operating conditions.

2. **Count-Rate Losses.** Calculation of the detector count rates for purposes of making dead time estimates requires that one calculate the *total* count rate, not only that due to U-235. Total count rate estimates for low-enrichment material must therefore take into account the relatively important background from U-238 gamma rays. If other radioactive materials are present within the sample, their contributions to the total count rate must also be considered.

Count-rate corrections can be made by determining the dead time or by making measurements for known live-time[5] intervals. The pile-up or overlap of electronic pulses is a problem which also results in a loss of counts in the full-energy peak for Ge(Li) systems. A pulser may be used to monitor and correct for these losses. Radiation which provides no useful information can be selectively attenuated by filters; e.g., a one-millimeter-thick cadmium filter will reduce x-ray interference, eliminating this source of count-rate losses.

3. **Instability in Detector Electronics.** The gain of a photomultiplier tube is sensitive to changes in temperature, count rate, and magnetic field. Provision can be made for gain checks and/or gain stabilization for enrichment measurement applications. Various gain stabilizers that automatically adjust the system gain to keep a reference peak centered between two preset energy limits are available.

C. REGULATORY POSITION

Passive gamma-ray spectrometry constitutes an acceptable means for nondestructively determining U-235 enrichment, if the following conditions are satisfied:

Range of Application

1. All material to be assayed under a certain calibration should be of similar chemical form, physical form, homogeneity, and impurity level.

2. The critical distance of the material should be determined. Only those items of the material having dimensions greater than this critical distance should be assayed by this technique.

3. The material should be homogeneous in all respects on a macroscopic[6] scale. The material should be homogeneous with respect to uranium enrichment on a microscopic[6] scale.

4. The containers should all be of similar size, geometry, and physical and chemical composition.

System Requirements

1. NaI(Tl) scintillation detectors having a resolution of FWHM $< 16\%$ at the 185.7-keV peak of U-235 are

[4] FWHM - full width of the spectrum peak at half its maximum height.

[5] "Live time" means 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

[6] Macroscopic refers to distances greater than the critical distance; microscopic to distances less than the critical distance.

generally adequate for measuring the enrichment of uranium containing more than the natural (0.71%) abundance of U-235. Crystals with a thickness of ~ 1.25 cm are recommended for optimum efficiency. If other radionuclides which emit significant quantities of gamma radiation in an energy region E = 185.7 keV ± 2 FWHM at 185.7 keV are present:

a. A higher-resolution detector, e.g., Ge(Li), should be used, or

b. A peak stripping procedure should be used to subtract the interference. In this case, data should be provided to show the range of concentration of the interfering radionuclide, and the accuracy and precision of the stripping technique over this range.

2. The detection system gain should be stabilized by monitoring a known reference peak.

3. The system should measure live time or provide a means of determining the count-rate losses based on the total counting rate.

4. Design of the system should allow reproducible positioning of the detector or item being assayed.

5. The system should be capable of determining the gamma-ray activity in at least two energy regions to allow background subtraction. One region should encompass 185.7 keV, and the other region should be above this but not overlapping. The threshold and width of the regions should be adjustable.

6. The system should have provisions for filtering low-energy radiation which could interfere with the 185.7-keV or background regions.

Data Reduction

1. If the total counting rate is determined primarily by the 185.7-keV gamma ray, the counting rate should be restricted (absorbers, decreased geometrical efficiency) below those rates requiring correction. The system sensitivity will be reduced by these measures and, if no longer adequate, separate calibrations should be made in two or more enrichment regions.

If the total counting rate is determined primarily by events other than those due to 185.7-keV gamma rays, counting rate corrections should be made.

2. To determine the location and width of the 185.7-keV peak region and the background region(s), the energy spectrum from each calibration standard (see Calibration, next section) should be determined and the position of the 185.7-keV peak and neighboring peaks noted. The threshold and width of each energy region should then be selected to avoid including any neighboring peaks, and to optimize the system stability and the signal-to-background ratio.

3. The net response attributed to 185.7-keV gamma rays should be the accumulated counts in the peak region minus a multiple of the counts accumulated in a nearby background region(s). A single upper background region may be monitored or both a region above the peak region and one below may be monitored.

If only an upper background region is monitored, the net response, R, should be given by

$$R = G - bB$$

where G and B are the gross counts in the peak region and the background region, respectively, and b is the multiple of the background to be subtracted. This net response, R, should then be proportional to the enrichment, E, given by

$$E = C_1 R = C_1 (G - bB)$$

where C_1 is a calibration constant to be determined (see Calibration, next section). The gross counts, G and B, should be measured for all the standards. The quantities G/E should then be plotted as a function of the quantities B/E and the slope of a straight line through the data determined. This slope is b, the multiple of the upper background region to be subtracted, i.e.,

$$G/E = b(B/E) + 1/C_1$$

The data from all the standards should be used in determining this slope.

If both an upper and a lower background are monitored, the counts in each of these regions should be used to determine a straight line fit to the background. Using this straight line approximation, the area or number of counts under this line in the peak region should be subtracted from the gross counts, G, to obtain the net response. An adequate technique based on this principle is described in the literature. [7]

Calibration [8]

1. Calibration standards should be obtained by:

a. Selecting items from the production material. A group of the items selected should, after determination

[7] G. Gunderson, I. Cohen, M. Zucker, "Proceedings: 13th Annual Meeting, Institute of Nuclear Materials Management," Boston, Mass. (1972) p. 221.

[8] None of the calibration techniques or data reduction procedures exclude the use of automated direct-readout systems for operation. The procedures described in this guide should be used for adjustment and calibration of direct-readout instruments.

of the gamma-ray response, be measured by an independent, more accurate technique traceable to, or calibrated with, NBS standard reference material, e.g., mass spectrometry. The other items should be retained as working standards.

b. Fabricating standards which represent the material to be assayed in chemical form, physical form, homogeneity, and impurity level. The U-235 enrichment of the material used in the fabrication of the standards should be determined by a technique traceable to, or calibrated with, NBS standard reference material, e.g., mass spectrometry.

2. The containers for the standards should have a geometry, dimensions, and composition which approximate the mean of these parameters in the containers to be assayed.

3. The values of enrichment for the calibration standards should span the range of values encountered in normal operation. No less than three separate standards should be used.

4. Each standard should be measured at a number of different locations, e.g., for a cylinder, at different heights and rotations about the axis. The mean of these values should be used as the response for that enrichment. The dispersion in these values should be used as an initial estimate of the error due to material and container inhomogeneity.

5. The data from the standards, i.e., the net response attributed to 185.7-keV gamma rays and the known uranium enrichment, should be used to determine the constants in a calibration function by a weighted least-squares technique.

Operations

1. The detection system and counting geometry (collimator and container-to-detector distance) should be identical to those used in calibration.

2. The data reduction technique and count-rate loss corrections, if included, should be identical to those used in calibration.

3. Data from all measurements should be recorded in an appropriate log book.

4. At least two working standards should be measured during each eight-hour operating shift. The measured response should be compared to the expected response (value used in calibration) to determine if the difference exceeds three times the expected standard deviation. If this threshold is exceeded, repeat measurements should be made to verify that the response is significantly different and that the system should be recalibrated.

5. All containers should be agitated, or the material mixed in some manner, if possible, prior to counting. One container from every ten should be measured at two different locations. Other items may be measured at only one position. (If containers are scanned to obtain an average enrichment, the degree of inhomogeneity should still be measured by this method.)

The difference between the measurements at different locations should be used to indicate a lack of the expected homogeneity. If the two responses differ by more than three times the expected standard deviation (which should include the effects of the usual or expected inhomogeneity), repeat measurements should be made to verify that an abnormal inhomogeneity exists. If the threshold is exceeded, the container should be rejected and investigated to determine the cause of the abnormal inhomogeneity. [9]

6. In the event that all containers are not filled to a uniform height, the container should be viewed at a position such that material fills the entire volume viewed by the detector. The procedure for determining the fill of the container should be recorded, e.g., by visual inspection at the time of filling and recording on the container tag.

7. The container wall thickness should be measured. The wall thickness and location of the measurement should be indicated, if individual wall thickness measurements are made, and the gamma-ray measurement made at this location. If the containers are nominally identical, an adequate sampling of these containers should be representative. The mean of the measurements on these samples constitutes an acceptable measured value of the wall thickness which may be applied to all containers of this type or category.

8. The energy spectrum from a process item selected at random should be used to determine the existence of unexpected interfering radiations and the approximate magnitude of the interference. The frequency of this test should be determined by the following guidelines:

a. At least one item in any new batch of material.
b. At least one item if any changes in the material processing occur.
c. At least one item per material balance period.

If an interference appears, either a higher-resolution detector must be acquired or an adequate peak stripping routine applied. In both cases additional standards which include the interfering radiations should be selected and the system recalibrated.

[9] The difference may also be due to a large variation in wall thickness.

9. No item should be assayed if the measured response exceeds that of the highest enrichment standard by more than twice the standard deviation in the response from this standard.

Error Analysis

1. A least-squares technique should be used to determine the uncertainty in the calibration constants.

2. The measurement-to-measurement error should be determined by periodically observing the net response from the standards and repeating measurements on selected process items. Each repeat measurement should be made at a different location on the container surface, at different times of the day, and under differing ambient conditions.[10] The standard deviation should be determined and any systematic trends corrected for.

[10] The statistical error due to counting (including background) and the errors due to inhomogeneity, ambient conditions, etc. will be included in this measurement-to-measurement error.

3. The item-to-item error due to the uncertainty in wall thickness should be determined. The uncertainty in the wall thickness may be the standard deviation about the mean computed from measurements on randomly selected samples, or it may be the uncertainty in the thickness measurement of individual containers. This uncertainty in wall thickness should be multiplied by the effect of a unit variation in wall thickness on the measured 185.7-keV response to determine this component uncertainty.

4. Item-to-item errors other that those measured, e.g., wall thickness, should be determined by periodically (see guidelines in paragraph 8. of the Operation Section) selecting an item and determining the enrichment by an independent technique traceable to, or calibrated with, NBS standard reference material. A recommended approach is to adequately sample and determine the U-235 enrichment by calibrated mass spectrometry. In addition to estimating the limit of error from these comparative measurements, the data should be added to the data used in the original calibration and new calibration constants determined.