



# REGULATORY GUIDE

OFFICE OF NUCLEAR REGULATORY RESEARCH

REGULATORY GUIDE 5.37  
(Task SG 047-4)

## IN SITU ASSAY OF ENRICHED URANIUM RESIDUAL HOLDUP

## A. INTRODUCTION

Part 70, "Domestic Licensing of Special Nuclear Material," of Title 10 of the Code of Federal Regulations requires licensees authorized to possess more than 350 grams of contained  $^{235}\text{U}$  to conduct a physical inventory of all special nuclear material in their possession at intervals not to exceed 12 months. Certain licensees authorized to possess more than one effective kilogram of special nuclear material are required to conduct more frequent measured physical inventories of their special nuclear materials. Further, these licensees are required to conduct their nuclear material physical inventories in compliance with specific requirements set forth in Part 70. Inventory procedures acceptable to the NRC staff are described in Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories."

Residual holdup is defined as the inventory component remaining in and about process equipment and handling areas after those collection areas have been prepared for inventory. In situ assay is used to ensure that a measured value of residual holdup is included in each material balance. This guide describes procedures acceptable to the NRC staff for the in situ assay of the residual enriched uranium holdup.<sup>1</sup> Because of the difficulty in measuring in-process holdup, the procedures described in this guide for calibration and error evaluation differ from general guidance on measuring residual holdup in more accessible controlled situations.

Any guidance in this document related to information collection activities has been cleared under OMB Clearance No. 3150-0009.

## B. DISCUSSION

Uranium accumulates in cracks, pores, and zones of poor circulation within and around process equipment. The

<sup>1</sup> Assay of residual plutonium holdup is the subject of Regulatory Guide 5.23, "In Situ Assay of Plutonium Residual Holdup." A proposed revision to this guide has been issued for comment as Task SG 045-4.

walls of process vessels and associated plumbing often become coated with uranium during processing of solutions. Uranium also accumulates in air filters and associated ductwork. Whenever possible, process equipment should be designed<sup>2</sup> and operated to minimize the amount of holdup. The absolute amounts of uranium holdup must be small for efficient processing and proper hazards control. However, the total holdup can be large (relative to the plant inventory difference (ID)) but have no significance on the ID if it remains reasonably constant. It is the change in the holdup between beginning inventory and ending inventory that may impact the inventory difference. The amounts of holdup must therefore be determined by an in situ assay.

Assay information can be used in one of two ways:

1. When the standard error of uranium holdup is compatible<sup>3</sup> with the plant standard error (estimator) of inventory difference (SEID), the material balance can be computed using the measured contents of uranium holdup. Additional cleanout and recovery for accountability will then not be necessary.

2. When the standard error of uranium holdup is not compatible with the plant SEID, the information obtained in the holdup survey can be used to locate principal uranium accumulations. Once located, substantial accumulations can be recovered, transforming the uranium to a more accurately measurable inventory component. Having reduced the amount of uranium holdup, the standard error

<sup>2</sup> Design features to minimize holdup in drying and fluidized bed operations, equipment for wet process operations, and equipment for dry process operations are the respective subjects of Regulatory Guides 5.8, 5.25, and 5.42.

<sup>3</sup> Compatibility exists when the contribution of the standard error of the holdup to the total plant SEID is not large enough to cause the overall SEID to exceed allowed limits. If the plant SEID exceeds allowed limits because of an excessive contribution from the holdup standard error, compatibility does not exist, and the remedial steps of paragraph 2 need to be taken.

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on the remeasurement of the remaining holdup may be sufficiently reduced to be compatible with overall plant SEID requirements.

The measurement procedures described in this guide involve the observation of the 185.7-keV gamma ray emitted in association with the decay of  $^{235}\text{U}$ . The amount of  $^{235}\text{U}$  holdup in a piece of equipment is proportional to the measured intensity of this gamma ray after suitable corrections are made (1) for attenuation by intervening materials, (2) for gamma ray self-attenuation by the uranium, (3) for geometrical factors that relate the measurement and calibration configurations, and (4) for background radiation from nearby sources of  $^{235}\text{U}$  not being measured. The proportionality factors are best determined prior to the holdup measurement by assays of known quantities of  $^{235}\text{U}$  distributed in well-defined and representative geometries as discussed below.

Note that there may be some cases in which gamma ray measurements with electronic detectors may not be practical or even feasible. Another gamma ray detection technique that might be attempted is thermoluminescence dosimetry (see References 1 and 2). This technique may be useful either as an independent check of a conventional gamma ray assay or as a substitute for it in environments or geometries where conventional detectors cannot be used. This technique will not, however, be discussed further in this guide.

The measurement of holdup in a complex plant environment can involve a very large number of measurements, as is implied in the text that follows. In a stable plant environment where the process behavior is well known and well characterized, it may be possible to arrange the holdup measurement program so that careful and extensive holdup measurements are made at infrequent intervals (for example, annually); at more frequent intervals (for example, at inventory times), careful measurements are again made in the (presumably fewer) known problem areas, and "spot check" measurements are made in the less problematic or less used zones where accumulations are known to be low. Such management of measurement resources can result in a very effective holdup measurement program at minimum cost.

## 1. DELINEATION OF COLLECTION ZONES

To accomplish the gamma ray assay, it is essential to consider the facility in terms of a series of zones that can be independently assayed. Such zones are designated "collection zones." Each uranium processing facility can be conceptually divided into a series of contiguous collection zones. Individual process machines, air filters, and separate item areas that can be isolated from one another may be suitable discrete collection zones. Great care needs to be taken to define all collection zones in such a way that (1) the assay of the zone can be performed with a minimum of interference and background from nearby zones (i.e., so that effective shadow shielding and reliable background subtraction can be accomplished); (2) the gamma ray detector can be positioned reproducibly and in such a way

that the gamma rays experience a minimum or easily predicted attenuation in the apparatus being measured; and (3) the distribution of material in the zone is represented, as much as possible, by one of the distribution geometries used in the calibration procedure (see Section B.4.1).

## 2. GAMMA RAY HOLDUP ASSAY

Two considerations are critical to the holdup assay. First, to perform an assay, the  $^{235}\text{U}$  gamma rays must reach the detector and be detected. Second, the observed response must be attributable to the collection zone being assayed. Therefore, the assay scheme is calibrated to compensate for the poor penetration of the  $^{235}\text{U}$  gamma ray. Also, the detector is collimated to separate a collection zone from its neighboring zones and from the radiation background. Finally, some effort may be necessary to employ external "shadow shielding" to block the field of view of the collimated detector from radiation being produced in collection zones other than the one being assayed.

For each gram of  $^{235}\text{U}$ , the 185.7-keV gamma ray is emitted at a rate of  $4.3 \times 10^4$  per second. This gamma ray is the only radiation emitted in the decay of  $^{235}\text{U}$  that is useful for this assay application. Unless mixed with plutonium or thorium, all other gamma rays are usually attributable to the Compton scattering of high-energy gamma rays emitted by the  $^{234}\text{Pa}$  daughter of  $^{238}\text{U}$ . The background at 185.7 keV due to this source of radiation varies depending on the length of time between the separation of the  $^{238}\text{U}$  daughters from the uranium (as frequently occurs during conversion processes) and the assay. This interference is very important for low-enrichment uranium but much less important at very high-enrichment values.

On uranium recycled after exposure in a nuclear reactor (where plutonium will be produced from the fertile  $^{238}\text{U}$ ), a sufficient quantity of  $^{232}\text{U}$  or  $^{237}\text{U}$  may be present to emit a measurable amount of interfering gamma radiation.

When uranium is mixed with thorium, the background gamma ray spectrum becomes much more complex. The background spectrum may vary because the daughters of  $^{232}\text{Th}$  can be volatilized to different extents during typical fuel processing. Further information on gamma ray interferences at energies near 185.7 keV may be found in Reference 3.

To ascertain total uranium holdup from the gamma ray assay of  $^{235}\text{U}$  holdup, it is necessary to determine from separate measurements the enrichment of the uranium holdup and to measure the emitted 185.7-keV gamma ray with sufficient resolution to enable the intensity of that gamma ray to be determined in the presence of the interfering radiations encountered. However, there are situations (for example, in multi-enrichment processing) in which holdup enrichment cannot be accurately measured because (1) the holdup enrichment may not be uniform and (2) the samples of holdup material may not be representative.

In such cases, the uncertainty in the total uranium holdup is increased drastically because of the uncertainty in the uranium enrichment used to convert the measured fissile holdup to total uranium holdup.

## 2.1 Gamma Ray Detection Instruments

Data processing electronics include a single-channel analyzer for the 185.7-keV photopeak, a timer-scaler unit, and a second single-channel analyzer used to determine the background radiation correction. Both detection channels get their signals from the same detector, and the signals are processed through the same electronics system. Battery-powered gamma ray analysis systems suitable for this application are commercially available and can enhance operational convenience. Methods for determining energy window settings are provided in References 4 and 5.

The detection efficiency and resolution of thallium-activated sodium iodide, NaI(Tl), detectors are generally adequate for this application when the uranium is not mixed with plutonium or thorium. Cadmium telluride (Refs. 6, 7, and 8) has better resolution than NaI and may prove adequate to resolve the 185.7-keV gamma ray from thorium or plutonium gamma rays. Lithium-drifted germanium, Ge(Li), and high-purity germanium, HPGe (also referred to as intrinsic germanium, IG), semiconductor gamma ray detectors have very high resolution but are less efficient than the other detector types and are more difficult to operate and maintain.<sup>4</sup>

Detector crystal dimensions are selected to provide a high probability of detecting the 185.7-keV gamma ray and a low probability of detecting higher energy radiation. For NaI, a crystal diameter of 1-1/2 in. (3.8 cm) and a thickness of 1/2 to 1 in. (1.3 to 2.5 cm) is recommended. For Ge(Li) and HPGe detectors, a planar crystal approximately 10 mm in depth is recommended.

## 2.2 Collimators and Absorbers for Gamma Rays

A shaped shield constructed of any heavy-element nonradioactive material is appropriate for gamma ray collimation. More than 98 percent of all 185.7-keV gamma rays striking a 0.35-cm-thick sheet of lead are absorbed or scattered.

The collimator will be most effective when it is concentric about the crystal and, for NaI detectors, the photomultiplier and its base. Extending the collimator forward of the crystal a distance equal to at least half the diameter of the crystal, and preferably the full diameter, is recommended (Refs. 4 and 5). Making this distance adjustable to reproducible settings will facilitate use of the collimated detectors for a range of collection zone sites. However, it is highly desirable to select collection zones

<sup>4</sup>For further information on germanium detectors, see Regulatory Guide 5.9, "Specifications for Ge(Li) Spectroscopy Systems for Material Protection Measurements," and references cited therein. A proposed revision to this guide has been issued for comment as Task SG 042-2 with the title "Guidelines for Germanium Spectroscopy Systems for Measurement of Special Nuclear Material."

and counting geometries so that one collimator setting will suffice for all measurements. This will simplify the calibration procedures since the calibration constants depend strongly on the collimator settings.

The detector collimator serves the purpose not only of defining the effective field of view but also of shielding the detector from unwanted radiation. To accomplish this latter purpose effectively, the collimator (heavy-element) material must also cover the rear of the detector as much as possible. This is usually easy to achieve with portable NaI detectors but requires more effort when Ge(Li) or HPGe detectors are used.

Often there is intensive X-ray radiation (in the energy range of 50 to 100 keV) coming from process equipment, and this radiation can tie up the detector electronics unnecessarily. To alleviate this problem, a 1/32-in.-thick (0.8-mm) layer of cadmium metal can be placed on the front face of the detector. This will absorb more than 90 percent of the (lower energy) X-rays incident upon the detector with a much smaller effect on the 185.7-keV gamma ray and will therefore render the detection system more sensitive in the energy region of interest.

## 2.3 Check Source for Gamma Ray Assay

It is important to check the operation of the detection system each time the instrumentation is moved or otherwise disturbed (e.g., power outage) during the course of each inventory sequence. An appropriate check source enables the stability of the assay instrument to be tested at any location. Such a source can be prepared by implanting a small encapsulated uranium source (containing about 0.5 gram of uranium) in the face of a plug of shielding material. The plug is shaped to fit and close the collimator channel, and the source is positioned adjacent to the crystal when the plug is in place. When the response from the check source remains within the expected value, the previous calibration data are assumed to be valid. If not, the energy window may have shifted, or the unit may be in need of repair and recalibration.

## 3. ISOLATION OF COLLECTION ZONES

To ensure that each collection zone is independently assayed, it is necessary to screen all radiations from the detector except those radiations emanating from the collection zone being assayed. This is principally accomplished through the use of the collimators described in Section B.2.2. Two additional means exist to further isolate a collection zone.

### 3.1 Detector Positioning

When uranium is located in back of the zone under assay in another collection zone or in a storage facility, the detector can be positioned between the radiation sources or above or below the collection zone to isolate the zone for assay.

## 3.2 Shadow Shielding

When it is not possible to avoid interfering radiations through the collimator design or through choosing the detector position for assay, it may be possible to move a shield panel between the source of interfering radiations and the collimator zone under assay. If the shield panel is sufficiently thick and its dimensions match or exceed the near portion of the collection zone under assay, no interfering radiations will penetrate through the shadow shield to the detector. A lead sheet 0.4 to 0.5 cm thick, which might be mounted on wheels as an upright panel, is generally adequate for this application.

## 4. CALIBRATION FOR HOLDUP MEASUREMENTS

### 4.1 Basic Counting Geometries

There are three fundamental counting geometries that can be used to represent most of the distributions of holdup in process equipment. These geometries are distinguished by the spatial distribution of the source material and the resulting dependence of the detector counting rate on the source-to-detector distance,  $r$ .

#### 4.1.1 Point Source

If the material being assayed is distributed over an area with dimensions that are small compared with the source-to-detector distance and if the material resides entirely within the detector field of view, the zone can be treated as a point source. The detector count rate for a point source varies inversely as the square of the source-to-detector distance (count rate is proportional to  $1/r^2$ ). Any equipment measured at great distances or any small pieces of equipment or equipment parts fall in this category. (Caution: small-sized sources of uranium could have very large self-attenuation of the 185.7-keV gamma ray and could therefore require great care in analysis.)

#### 4.1.2 Line Source

If the material being assayed is evenly distributed along a linear path so that only a segment of that distribution length is contained in the detector field of view, this material can be treated as a line source. The detector count rate for a line source varies inversely as the source-to-detector distance (count rate is proportional to  $1/r$ ). Examples of this type of holdup geometry include isolated sections of piping and long, narrow trays or columns.

#### 4.1.3 Area Source

If the material being assayed is spread over an area large enough for the material to cover the full field of view of the detector for a range of source-to-detector distances, the material can be assayed as an area source. As long as the material being viewed is uniformly distributed, the detector count rate will be independent of the source-to-detector distance. However, for holdup applications, uniform material distribution is rare; therefore, the source-to-detector distance can affect the instrument response and

needs to be specified. It should be further noted that, when there are several measurement locations covering a large area (such as a floor), it is important to maintain the same source-to-detector distance (even if material distribution is uniform within a given measurement area) so that the number of measurement areas needed to cover the entire area remains constant. Examples of this type of assay geometry include floors, walls, glovebox floors, and any large-area pieces of equipment.

### 4.2 Calibration of Detector Response

#### 4.2.1 Mockup of Known Material Distributions

For a given collimator setting, the detector response for the three basic source distribution geometries listed above needs to be determined. For the point source, the response is expressed as (counts per minute)/gram of  $^{235}\text{U}$  at a specified source-to-detector distance. For the line source, the response is expressed as (counts per minute)/(gram of  $^{235}\text{U}$  per unit length) at a specified source-to-detector distance. For the area source, the response is expressed as (counts per minute)/(gram of  $^{235}\text{U}$  per unit area) at a specified source-to-detector distance. Corrections to the point and line source calibrations for different detector distances in the actual holdup measurements are made using their  $1/r^2$  and  $1/r$  count-rate dependences, respectively. For further detailed discussion of the measurement of detector responses for these basic geometries, see Reference 9.

The measurement of the point source response can be accomplished with an encapsulated uranium foil smaller in size than the detector collimator opening. This foil can also serve as the check source for verification of the continued stability of the instrument settings in the field.<sup>5</sup> Care must be taken in the preparation of this calibration standard to ensure that the amount of  $^{235}\text{U}$  is well known. It is also important to measure the gamma ray attenuation through the encapsulating material and the self-attenuation of the uranium foil and to subsequently correct the calibration standard response to compensate for these effects. Enough  $^{235}\text{U}$  needs to be present in this standard to provide count rates that will ensure good statistical precision of the calibration in a reasonable period of time.

The measurement of the line source response is best accomplished by constructing a cylindrical surface distribution of  $^{235}\text{U}$  with the aid of large uranium foils rolled into a cylindrical shape. (It is also possible to establish the line source response using a point source, as described in Reference 5). The line source geometry is closest to that of the pipes and ducts likely to be encountered in actual measurements. The amount of  $^{235}\text{U}$  in the foils must be well known to ensure an accurate calibration. The area source response can also be measured with the same uranium foils laid flat to simulate the expected uranium distribution on surfaces such as walls and floors. Self-attenuation by the foils must also be taken into account in order for the calibration to be as accurate as possible.

<sup>5</sup>Note that a calibration source can be used as a check source, but a check source cannot be used as a calibration source.

There may be special material distribution geometries in the facility that are not readily represented by one of the three basic configurations described above. These special geometries may be mocked up as carefully as possible with large uranium foils and point sources to produce a usable detector response calibration for these special cases. Examples of special cases might be concave or convex equipment surfaces or the internal volume of a rectangular cavity (see Reference 9). Because material particle sizes (or material deposit thicknesses) have a significant effect on the self-attenuation of the gamma ray signals, it is important to use (whenever practical) well-characterized process material for preparing calibration standards and to duplicate to the extent possible process holdup distribution relative to particle size or thickness. Furthermore, holdup in floors is often deposited at various depths into the floor, rather than on the surface. Thus, calibration standards for such measurements need to incorporate the appropriate geometry and matrix effects. Core samples of a floor may be needed to establish typical concentrations at various floor depths.

Calibration of the holdup measurement system using this procedure is recommended until a history of comparisons between predicted and recovered holdup quantities is developed. If it is possible to take holdup measurements before and after the cleanout of a piece of shut-down process equipment, these can be used to establish this history of comparisons and improve the accuracy of the calibration for each collection zone.

#### *4.2.2 Measurement of Check Sources in Actual Process Equipment*

One method for calibrating detector response to holdup radiation in process equipment is to place a known calibration source in various positions in that equipment and record the detector responses. In this way, the overall detector response (including all corrections for attenuation and geometry) is determined empirically. Unfortunately, this procedure is impractical, if not impossible, in process equipment already in operation. However, those responsible for holdup assays need to be aware of occasions when new process equipment is brought into the plant for installation. At that time, before installation, calibration sources can conveniently be placed in the equipment and the empirical measurements of the detector responses can be made. This procedure would be a valuable supplement to calibration data obtained from mockups of standard counting geometries and comparisons with cleanout recovery data.

## **5. HOLDUP MEASUREMENTS AND STANDARD ERROR**

### **5.1 Assay Measurements**

In performing the holdup measurements, one must be aware of the large uncertainties in holdup assays arising primarily from variability in the measurement conditions (e.g., background, geometry, gamma ray attenuation, material distribution). Accordingly, every effort should be made to perform the assays from as many vantage points as possible for each collection zone. If this is impractical on a

routine basis because of time or space constraints, one might consider multiple measurements on a collection zone initially followed by fewer routine measurements at representative assay sites. Careful thought in selecting measurement points and measurement strategy will minimize ambiguities in the interpretation of the data.

#### *5.1.1 Detector Positioning*

Location and configuration of collection zones are established on the basis of a detailed physical examination and a radiation survey of the physical layout of the facility. Preliminary measurements are needed to determine the optimum detector positions for the holdup assays. If nonuniform distribution of material in a collection zone is suspected or if the process apparatus is sufficiently complicated to require extensive attenuation corrections for certain counting geometries, multiple measurements are advisable for that collection zone and more than one detector position may be necessary. If radiation surveys have indicated zones of high holdup, extra care will be necessary in the holdup measurements for these zones to minimize the contribution of their holdup uncertainties to the SEID. Selecting optimum detector positions includes consideration of the need to conveniently measure the line-of-sight background by moving the detector to one side without changing its orientation.

#### *5.1.2 Holdup Measurements*

The measurement and analysis of the 185.7-keV gamma ray intensity from a collection zone may be carried out by treating the material distribution as one of the three basic geometries described in Section B.4.1 or as one of the special cases that may have been measured, as mentioned in Section B.4.2. If the nature of the material distribution is uncertain for a particular detector position, a measurement of the detector counting-rate dependence on the source-to-detector distance,  $r$ , may indicate the most appropriate counting-rate geometry with which to interpret the data.

After the assay positions for the detector and shadow shields are established for each collection zone, permanent markings that indicate detector location (including height) and orientation will ensure reproducibility of subsequent measurements for these positions. Uniquely labeling each assay site will facilitate unambiguous reference to each measurement and its location in the assay log.

After measuring the 185.7-keV gamma ray intensity at each detector position in a given collection zone, the line-of-sight background is measured by moving the detector and collimator to one side of the zone (still pointing in the same direction as during the assay) and counting the 185.7-keV gamma ray intensity from the surrounding materials. During the background measurement, the vessel in which the holdup is being measured must not be in the field of view of the detector. This procedure is repeated at all measurement positions and in all counting geometries designated for each collection zone. The final holdup value for the zone is obtained from the average of the individual

measurements (each one being corrected for the effects of attenuation and any variation in geometry relative to the calibration measurement).

Whenever possible, the collection zone is assayed in a variety of ways. For example, one could measure an apparatus at close range, treating it as an area source, and then repeat the measurement at long range, treating the zone as a point source. It may be better to measure some zones from several different directions—especially if complicated attenuation corrections are called for in some of the counting geometries. The averaging of several independent measurements of one zone helps both to smooth out imprecisions due to incomplete knowledge of the measurement conditions and to provide a measure of the magnitudes of the fluctuations in the measurement results as an estimate of the measurement variability.

### 5.1.3 Gamma Ray Attenuation Corrections

To obtain useful assay results by detecting the 185.7-keV gamma ray, it is necessary to correct each assay for attenuation of the signal, either within the uranium holdup material or by structural materials. Without this critical correction, the assay is no more than a lower limit on the true holdup value. Details for establishing an appropriate attenuation correction are given in Laboratory Exercise No. 4 of Reference 5. Additional treatment of gamma ray attenuation corrections is given in Reference 10.

### 5.1.4 Uranium Enrichment

The 185.7-keV gamma ray measurement provides the amount of  $^{235}\text{U}$  holdup. Total uranium holdup is obtained by dividing the assay result by the declared material enrichment in  $^{235}\text{U}$ . For accountability of fissile uranium, this step is not necessary. However, as is discussed below, conversion of the measurement results to total uranium is necessary when making comparisons with other assay techniques that use elemental analysis. Also note that, at low enrichment (and to a lesser extent at high enrichment), some knowledge of the total amount of uranium holdup is necessary to facilitate proper gamma ray self-attenuation corrections to the measurements.

## 5.2 Assignment of Standard Error

The assignment of a standard error (i.e., twice the standard deviation,  $\sigma$ ) to a holdup measurement is extremely difficult on a rigid statistical basis. This situation exists because the only statistically predictable fluctuations (i.e., counting statistics) in this application are frequently negligible compared with the variability due to counting geometry (including material distribution), gamma ray attenuation, gamma ray background and interferences, and instrument instabilities. Therefore, the variability can be large and it is necessary to guard against underestimating the standard deviation of the overall holdup value in a collection zone. Careful measurements are needed during the calibration procedure to determine the range of detector responses resulting from variations in measurement parameters. A useful discussion of these ideas is presented in Reference 9.

A reasonable estimate of the uncertainty in the measured holdup for a given collection zone may be obtained by considering the range of holdup values obtained from the variety of measurements performed on that collection zone as suggested in the previous section. The mean value for the holdup is defined as the average of the various (corrected) measurement results on the collection zone. In view of the well-known uncertainty of holdup measurement, the standard deviation for that mean value is conservatively estimated as one-half of the range of holdup values obtained in the measurements.

In some cases, counting statistics may be so poor that they contribute significantly to the measurement variability. In such an instance, the standard deviation of the overall holdup  $\sigma_{(h-u)}$  is defined as the square root of the sum of the squares of the standard deviation due to counting  $\sigma_{(stat)}$  standard deviation due to measurement fluctuations  $\sigma_{(meas)}$ ; that is,

$$\sigma_{(h-u)} = \sqrt{\sigma_{(stat)}^2 + \sigma_{(meas)}^2} \quad (1)$$

## 5.3 Estimation of Bias

When a single collection zone is cleaned out, it is desirable to perform a holdup assay before ( $H_{\text{before}}$ ) and after ( $H_{\text{after}}$ ) the cleanout if possible. By comparing the amount of uranium removed,  $U_r$ , to the amount predicted through the in situ holdup assays,  $U_a$ , the collection zone calibration can be updated, and the calibration and assay standard deviations can be based on relevant data. The amount of uranium recovered,  $U_r$ , during the cleanout of a specific collection zone can be assayed through sampling and chemical analysis or through other applicable nondestructive assay methods.<sup>6</sup>

The update data is computed as the difference in the holdup assays before and after the cleanout:

$$U_a = H_{\text{before}} - H_{\text{after}} \quad (2)$$

The percentage difference between the assay and recovery values for the uranium holdup,

$$\Delta = 100(U_a - U_r)/U_r \quad (3)$$

is then computed. A running tabulation of the quantities  $U_a$ ,  $U_r$ , and  $\Delta$  (as well as their standard deviations,  $\sigma_a$ ,  $\sigma_r$ , and  $\sigma_\Delta$ ) is kept in the assay log for each collection zone.

The average value,  $\bar{\Delta}$ , of the percentage differences between  $U_a$  and  $U_r$  will serve as an estimate of the bias in the holdup assay for that collection zone and will also provide quantitative justification for revision of the assay calibration for that zone to remove the bias. The root-mean-square deviations,  $s_{\bar{\Delta}}$ , of the percentage differences,

<sup>6</sup>See, for example, Regulatory Guide 5.11, "Nondestructive Assay of Special Nuclear Material Contained in Scrap and Waste." A proposed revision to this guide has been issued for comment as Task SG 043-4.

$\Delta_i$ , from their mean value,  $\bar{\Delta}$ , serves as a check on the appropriateness of the size of the estimated standard deviations of the holdup measurements. To the extent that the standard deviation of  $U_r$  is small compared with the standard deviation of  $U_a$  (usually an adequate assumption), the quantity  $s_{\Delta}$  should be comparable in size to the standard deviation of  $U_a$ . For  $K$  measurements of the percentage differences  $\Delta_i$  for a given collection zone, the quantity  $s_{\Delta}$  is given by:

$$s_{\Delta} = \left[ \frac{\sum_{i=1}^K (\Delta_i - \bar{\Delta})^2}{(K-1)} \right]^{1/2} \quad (4)$$

Equation 4 assumes that all of the  $\sigma_{\Delta}$ 's are equal. For a calculation of  $s_{\Delta}$  using weighted sums, see Reference 11.

Note that, if the holdup measurements (i.e.,  $H_{\text{before}}$  or  $H_{\text{after}}$ ) contain a constant bias, their difference can still provide useful information in the comparison with  $U_r$ . However, a small difference between  $U_a$  and  $U_r$  does not necessarily mean that the error associated with  $H$  is small. This ambiguity is reduced in importance if the cleanout is such that  $H_{\text{after}}$  is much smaller than  $H_{\text{before}}$ . In addition, the use of several holdup measurements from various vantage points, as suggested earlier, will help to minimize the bias associated with incorrect geometrical or attenuation corrections in one measurement configuration.

#### 5.4 Effect of Enrichment Uncertainty

The gamma ray measurements described here provide a direct determination of the fissile uranium (i.e.,  $^{235}\text{U}$ ) holdup in the zone under consideration. However, the comparison and verification measurements made with chemical techniques provide elemental analysis without consideration of the isotopic makeup of the samples. While  $^{235}\text{U}$  accountability does not require it, knowledge of the uranium enrichment of the material being measured is necessary for meaningful comparisons with the chemical analyses. Thus, the holdup assays must be divided by the material enrichment to make the comparisons outlined in Equations 2 through 4, which are expressed in terms of total uranium.

If the process equipment is thoroughly cleaned each time the enrichment of the uranium feed is changed, the holdup will consist primarily of the current material. In that case, the declared enrichment can be used. When mixing occurs, use of the stream-averaged enrichment is appropriate. The enrichment uncertainty bounds are estimated by considering the batches of highest and lowest enrichment and computing the corresponding range. The uncertainty in the material enrichment must then be incorporated into the quoted holdup uncertainty before making direct comparisons with the chemical analyses.

### C. REGULATORY POSITION

To develop a program acceptable to the NRC staff for the periodic in situ assay of enriched uranium residual

holdup as a method for measuring this inventory component, the factors in the following sections should be considered.

## 1. DELINEATION OF COLLECTION ZONES AND ASSAY SITES

Preliminary radiation survey measurements of the uranium processing facility should be used to establish independent collection zones and detector positions within the zones.

At each collection zone, detector positions (assay sites) should be chosen so that the material holdup can be measured from several vantage points around the zone. At each assay site, the detector should have exclusive view of the collection zone being assayed. If necessary, shadow shielding should be used to isolate the region being assayed from other collection zones. Detector positions should be chosen to minimize the measurement ambiguities, as described in Section B.5.1.1.

Each assay site should be permanently marked with paint or colored tape on the floor to ensure reproducible assay positions. Detector height and orientation should be clearly indicated in the assay log for each measurement site and, if possible, included with the site markings. The markings should be protected (for example, with clear epoxy) to ensure their long-term durability.

Each assay site should be uniquely labeled to facilitate unambiguous reference to that site in the assay log.

Areas may be denoted as "problem areas" so that careful holdup measurements will be made there each time plant holdup is to be determined, or an area may be labeled as a "spot check" zone where accumulations are known to be low and careful holdup assays are needed less frequently.

## 2. ASSAY SYSTEM

### 2.1 Detector Selection

NaI(Tl) detectors are generally suitable for this application when the uranium is not mixed with thorium or plutonium. The crystal depth should be sufficient to detect a significant percentage of 185.7-keV gamma rays. For NaI(Tl), a 1-inch (2.5-cm) crystal depth is recommended. Cadmium telluride, Ge(Li), or HPGe detectors should be used when NaI resolution is inadequate to separate the  $^{235}\text{U}$  activity from interfering radiations.

The crystal should be stabilized with a suitable radioactive source. An internal seed containing  $^{241}\text{Am}$  is recommended for NaI applications. The electronics should be capable of stabilizing on the reference peak produced by the seed.

Two single-channel analyzers should be provided with lock-set energy windows. These analyzers should be integrally packaged, should get their signals from the same detector, and should process the signals with the same

electronics. One channel should be set to admit the 185.7-keV gamma rays from  $^{235}\text{U}$ . The second channel should be set above the first window to provide a background correction for the assay window. The electronics unit should have a temperature stability of less than 0.1 percent per degree Celsius.

## 2.2 Gamma Ray Collimator

A cylinder of shielding material such as lead should be made coaxial with the gamma ray detector. The end of the cylinder opposite the crystal should be blocked with the shielding material. The thickness of the collimator should be chosen to provide sufficient directionality for the specific application (a lead thickness of 0.35 cm should be sufficient for most applications). The collimator shield should be fixed over the end of the detector crystal at a reproducible setting identical to that used in the calibration measurements.

An absorber should be placed over the front face of the detector to filter out unwanted low-energy photons. A 1/32-in.-thick (0.8-mm) layer of cadmium metal is recommended.

## 2.3 Gamma Ray Calibration and Check Sources

Standard sources of  $^{235}\text{U}$  should be provided for calibrating the measurement system for the basic measurement geometries described in Section B.4. A small encapsulated foil of enriched uranium can be used both as a calibration standard for the point-source counting geometry and as a check source for verification of instrument stability.<sup>7</sup> Standard sources for the line and area material distributions should be well-characterized uranium samples such as large-area uranium foils of well-known thickness and  $^{235}\text{U}$  content, which can be arranged in reproducible configurations for the calibration measurements. Other well-characterized samples taken from the process may also be advisable for use as calibration standards to reflect more realistically actual process conditions. The gamma ray self-attenuation correction (or equivalently, the effective mass of  $^{235}\text{U}$  without application of the correction) should be clearly specified for all of the uranium foils and check sources.

## 3. CALIBRATION

### 3.1 Instrument Check

The stability of the gamma ray detection system should be tested prior to each inventory. If the check-source measurement is consistent with previous data (i.e., is within plus or minus two single-measurement standard deviations of the mean value of previous data), previously established calibration data should be considered valid. If the measurement is not consistent, the operation of the unit should be

<sup>7</sup>Recall that a calibration source may be used as a check source, but a check source should never be used for calibration.

checked against the manufacturer's recommendations and repaired or recalibrated as required.

### 3.2 System-Response Calibration

The response of the detection system should be determined with well-known quantities of  $^{235}\text{U}$  in the basic measurement geometries described in Section B.4. If there are special counting geometries in the facility that are not readily represented by one of the basic configurations, these geometries should also be mocked up and measured during the calibration procedure.

## 4. ASSAY PROCEDURES

### 4.1 Assay Log

An assay log should be maintained. Each collection zone or subzone should have a separate section in the assay log, with the corresponding calibration derived on the page facing the assay data sheet. Recording space should be provided for the date of measurement, gross counts, corrected counts, and the corresponding grams of uranium from the calibration in addition to verifying the position and electronic setting of the instrument. Also, space in the log should be provided for recording data from recovery operations and holdup assay comparisons as described in Section B.5.3.

### 4.2 Preassay Procedures

Prior to inventory, the enrichment of the uranium processed during the current operational period should be determined. Variations in the gamma ray yield data from the calibration standard should be calculated. Either the calibration data or the predicted holdup should then be corrected to reflect this change.

Before each inventory, the operation of the gamma ray assay detection systems should be checked, as described in Regulatory Position 3.1.

Prior to any assay measurements, feed into the process line should be stopped. All in-process material should be processed through to forms amenable to accurate accountability. All process, scrap, and waste items containing uranium should be removed to approved storage areas to minimize background radiations.

### 4.3 Measurements

Before beginning the holdup measurements, it is advisable to conduct a preliminary gamma survey of the collection zones to point up the zones where holdup accumulations are the highest (and therefore where the most careful measurements should be made). In zones where accumulations are shown to be very low by the survey, spot check measurements may be adequate, as pointed out earlier.

Before assaying each collection zone, the operator should verify that floor location, probe height, probe

orientation, and electronics settings correspond to previous measurements. All check and calibration sources should be sufficiently removed (or shielded) to prevent interference with the measurement. Prior to taking a measurement, a visual check of the zone and the line of sight of the detector probe should be made to ensure that no obvious changes have been made to the process area and that no unintended accumulations of uranium remain within the collection zone. The operator should initial the measurement log to ensure compliance for each collection zone.

When all the preceding steps have been completed, the measurements at each collection zone should be taken and recorded. An attenuation correction measurement should be made, and the corrected response should be converted to grams of  $^{235}\text{U}$  and recorded in the assay log. To convert the result to grams of uranium, divide the previous result by the declared uranium enrichment. If a high response is noted, the cause should be investigated. If the collection zone contains an unexpectedly large content of uranium, that collection zone should be cleaned to remove the accumulation for conversion to a more accurately accountable material category. After the cleanout has been completed, the zone should be reassayed and the assay difference before and after cleanout should be compared

with the recovered material quantity to test the validity of the zone calibration as described in Section B.5.3.

## 5. ESTIMATION OF HOLDUP UNCERTAINTY

During the initial implementation of the holdup measurement program, the holdup uncertainty for each collection zone should be estimated from the range of values obtained in the various measurements on that zone as described in Section B.5.2. As a history of comparisons between holdup measurements and cleanout recovery data becomes available, these data should be used to adjust for bias and to revise the magnitudes of the holdup uncertainties as described in Section B.5.3.

During each physical inventory, the calibration in at least 10 percent of the collection zones should be updated on the basis of the comparison between holdup and cleanout recovery measurements. In small plants with less than ten collection zones, at least one zone should be updated during each physical inventory.

To ensure that error predictions remain current, data from only the 12 preceding independent tests should be used to estimate the assay uncertainty.

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## VALUE/IMPACT STATEMENT

### 1. PROPOSED ACTION

#### 1.1 Description and Need

Regulatory Guide 5.37 was published in August 1974. The proposed action, a revision to this guide, is needed to bring the guide up to date with respect to advances in measurement methods and changes in terminology.

#### 1.2 Value/Impact Assessment

##### 1.2.1 NRC Operations

The regulatory positions will be brought up to date.

##### 1.2.2 Other Government Agencies

Not applicable.

##### 1.2.3 Industry

Since industry is already applying the methods and procedures discussed in the guide, updating these should have no adverse impact.

##### 1.2.4 Public

No adverse impact on the public can be foreseen.

#### 1.3 Decision on the Proposed Action

Revised guidance should be developed to reflect the improvement in measurement techniques and to bring the language into conformity with current usage.

### 2. TECHNICAL APPROACH

Not applicable.

### 3. PROCEDURAL APPROACH

Of the procedural alternatives considered, revision of the existing regulatory guide was selected as the most advantageous and cost effective.

### 4. STATUTORY CONSIDERATIONS

#### 4.1 NRC Authority

Authority for the proposed action is derived from the Atomic Energy Act of 1954, as amended, and the Energy Reorganization Act of 1974, as amended, as implemented through the Commission's regulations.

#### 4.2 Need for NEPA Assessment

The proposed action is not a major action that may significantly affect the quality of the human environment and does not require an environmental impact statement.

### 5. RELATIONSHIP TO OTHER EXISTING OR PROPOSED REGULATIONS OR POLICIES

The proposed action is one of a series of revisions of existing regulatory guides on nondestructive assay techniques.

### 6. SUMMARY AND CONCLUSIONS

A revised guide should be prepared to bring Regulatory Guide 5.37 up to date.

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