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PROPOSED REVISION 1* TO REGULATORY GUIDE 5.23
IN SITU ASSAY OF PLUTONIUM 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 1 kilogram of plutonium to calculate a material balance based on a measured physical inventory at intervals not to exceed 2 months. 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 detailed in Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories."

Plutonium residual holdup is defined as the plutonium inventory component remaining in and about process equipment and handling areas after these collection areas have been prepared for inventory. Whenever possible, process equipment should be designed** and operated so as to minimize the amount of holdup. In this guide, procedures for the in situ assay of the plutonium residual holdup are described.

Assay information may be used in one of two ways:

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* The substantial number of changes in this proposed revision has made it impractical to indicate the changes with lines in the margin.

** Design features to minimize holdup in process equipment are the subject of a series of regulatory guides (5.8, 5.25, and 5.42).

1. When the limit of error of plutonium holdup is compatible* with constraints on the overall limit of error on the facility inventory difference (LEID), the material balance can be computed using the measured contents of plutonium holdup. Additional cleanout and recovery for accountability will then not be necessary.

2. When the limit of error of plutonium holdup is not compatible with constraints on the overall LEID, the information obtained in the holdup survey can be used to locate principal plutonium accumulations and to ensure that other areas of the process contain less than the detectable amount of plutonium. Once located, substantial accumulations can be recovered, transforming the plutonium to a more accurately measurable inventory component. Having reduced the amount of plutonium holdup, the limit of error on the remeasurement of the remaining holdup may be sufficiently reduced to be compatible with overall LEID requirements.

B. DISCUSSION

Plutonium accumulates in cracks, pores, and zones of poor circulation within process equipment. The walls of process vessels and associated plumbing often become coated with plutonium during solution processing. Surfaces internal and adjacent to process equipment, especially glovebox walls and floors, accumulate deposits of plutonium that can become appreciable. Plutonium also accumulates in air filters and associated ductwork. The absolute amounts of plutonium 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 significant impact 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 ID.

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*When the contribution of the limit of error of the holdup to the total plant LEID is not large enough to cause the overall LEID to exceed allowed limits, the compatibility criterion is satisfied. If the plant LEID exceeds allowed limits because of an excessive contribution from the holdup limit of error, the compatibility criterion is not satisfied, and remedial steps should be taken in accordance with paragraph 2.

The measurement procedures described in this guide involve the detection of gamma rays and neutrons that are spontaneously emitted by the plutonium isotopes. Because the gamma rays of interest are emitted by the major isotope, Pu-239, gamma ray assay is the preferred method whenever its acceptance criteria are satisfied. The amount of Pu-239 holdup in a piece of equipment is proportional to the measured intensity of the emitted gamma rays after suitable corrections are made for attenuation by intervening materials, for self-attenuation by plutonium, for geometrical factors, and for background radiation.

If plutonium is held up in furnaces, grinders, or other heavy equipment that is too dense to permit the escape of gamma rays, an assay based on spontaneous fission neutrons from the even isotopes of plutonium may be possible. This technique requires knowledge of the isotopic composition of the plutonium and some knowledge of its chemical form.

Thermoluminescent dosimetry is a third technique that can be used to measure holdup from the inside of large pieces of equipment. This technique is also useful for carrying out measurements in an unobtrusive manner outside normal plant operating hours. For further details on this technique, see Section B.2.3 of this guide.

For all three techniques, the proportionality factors between amount of holdup and detector response are best determined prior to the holdup measurement by assays of known quantities of plutonium distributed in well-defined and representative geometries, as discussed below.

1. DELINEATION OF COLLECTION ZONES

Typical plutonium process facilities comprise a number of interconnected gloveboxes that contain work areas and most process equipment, in-process storage areas, and self-contained process equipment. Also, solution processing requires tanks, plumbing, and pumping equipment, which are often located in close proximity to the glovebox lines. Finally, storage areas for feed, scrap and waste, and final product are often located in close proximity to the plutonium process area.

To accomplish the holdup measurements, 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 plutonium-processing

facility can be conceptually divided into a series of contiguous collection zones on the basis of process activities and collection geometries. Individual machines, filters, pipes, tanks, gloveboxes, or surface areas that can be isolated from one another may be suitable discrete collection zones. Great care should be taken to define all collection zones so that (1) the assay of the zone can be performed with a minimum of interference from nearby zones, (2) the detector can be positioned reproducibly and in such a way that the radiation being measured experiences a minimum, or easily predicted, attenuation in the apparatus being measured, and (3) the distribution of material in the zone can be represented by one of the distribution geometries used in the calibration procedure described below.

Gamma ray assay for plutonium holdup is practical when the collection zone consists of a single structure of relatively uniform cross section. When a collection zone contains a complex item of equipment with significant self-shielding properties, the uncertainty in the holdup measurement may be primarily due to attenuation of radiation in the internal structure. In such cases, neutron assay from the outside and thermoluminescent dosimeter assay from the inside may be applicable.

2. APPLICABLE METHODS AND INSTRUMENTS

Two considerations are critical to the selection of methods and instruments. First, to perform an assay, one must ensure that the plutonium radiations reach the detector and are detected. Second, the observed response must be attributable to the collection zone being assayed. Therefore, the assay scheme is developed around penetrating radiations, and the detector is collimated to provide for sufficient directionality in the response to resolve a collection zone from its neighboring zones and from the background. Finally, some effort may be necessary to employ external "shadow shielding" to block radiation being produced in adjacent collection zones from the field of view of the collimated detector.

2.1 Gamma Ray Assay

Under closely controlled conditions, the measured plutonium gamma ray spectrum can be interpreted in terms of the abundance of each gamma ray

emitter present in the sample. Because of the large number of gamma rays (Refs. 1 and 2) present, many regions of the observed spectrum are characterized by overlapping lines. To accomplish the assay, it is necessary to select an appropriate spectral region and provide a detection system with sufficient resolution to measure the activity from the isotopes of interest.

Gamma ray assay has an advantage over neutron assay in that the emissions are primarily from the principal isotopes of interest. Because of the high emisson rate of gamma rays, a detection sensitivity of less than 1 gram is generally attainable.

The most useful portion of the spectrum for holdup assay is the Pu-239 gamma ray complex in the 375 to 450 keV range.* The yields of these lines are given in Table 1.

Table 1

PROMINENT GAMMA RAYS FROM Pu-239 IN 375-450 keV ENERGY RANGE

Energy	Intensity (γ/sec-g Pu-239)
375.0	3.59×10^4
380.2	0.70×10^4
382.7	0.59×10^4
392.5	0.26×10^4
393.1	1.01×10^4
413.7	3.43×10^4
422.6	0.27×10^4
Total	9.85×10^4

* In typical NaI detectors (with energy resolution of 10 percent at 414 keV), the 414-keV photopeak will produce counts in the approximate energy range of 373 to 455 keV. Thus, an energy window from 375 to 450 keV will include most of the 414-keV full-energy counts for a variety of detector resolutions. Furthermore, such a window setting will include a significant fraction of the 375-keV Pu-239 gamma rays (see Table 1), but will also exclude most of the potentially interfering 332-keV gamma rays from Am-241 or Pu-241.

2.1.1 Gamma Ray Detection Instruments

Gamma ray detection systems consist of a scintillation or semiconductor gamma ray detector and appropriate electronics (Refs. 3 and 4). Required electronics include at least a single-channel analyzer and a timer-scaler unit. A second single-channel analyzer viewing the same detector pulses used to determine the background radiation correction is a timesaving feature. A number of portable battery-powered systems are commercially available for this application.

The detection efficiency and resolution (10-15 percent) of NaI(Tl) is generally adequate for holdup measurements. CdTe, Ge(Li), and intrinsic germanium detectors have better resolution than NaI(Tl) but are more costly and more difficult to operate. For more information on Ge(Li) and intrinsic germanium detectors, see Regulatory Guide 5.9, "Guidelines for Ge(Li) Spectroscopy Systems for Material Protection Measurements,"* and the references cited therein.

The 332.3-keV gamma ray from U-237, a short-lived (6.75 d) daughter of Pu-241, is usually the principal interference for Pu-239 assay by NaI detection of the 375 to 450 keV complex. If the U-237 is in equilibrium with Pu-241, the intensity of this gamma ray is 1.15×10^6 ρ/sec-g Pu-241. Since this gamma ray is also emitted in the decay of Am-241, the interference from this decay branch may also be important in case of preferential americium holdups. To avoid this interference when using NaI detectors, the assay energy window is adjusted to span the range from 390 to 450 keV for plutonium holdup with high americium content.

Detector crystal dimensions are selected to provide a high probability of detecting gamma rays from the 375 to 450 keV complex and a low probability of detecting high-energy radiation. For NaI, a crystal diameter of 2 inches with a thickness of 2 inches is recommended. For germanium detectors, a moderate-volume coaxial detector is recommended.

2.1.2 Collimators and Absorbers for Gamma Rays

A shaped shield constructed of any heavy-element material is appropriate for gamma ray collimation. For cost, availability, and ease of fabrication,

*A proposed revision to this guide has been issued for comment as Task SG 042-2.

lead is recommended. Less than 2 percent of all 400-keV gamma rays striking a 1.5-cm-thick sheet of lead will pass through without suffering an energy loss.

The collimator will be most effective when it is concentric about the crystal and photomultiplier and completely covers the photomultiplier base. Extending the collimator forward of the crystal at least a distance equal to half the diameter of the crystal, and preferably the full diameter, is recommended (Ref. 5). Making this distance variable to reproducible settings will permit adjustment over a range of collection zone sizes. However, every effort should be made to select collection zones and counting geometries so that one collimator setting will suffice for all measurements. This will simplify the calibration procedures because the calibration constants depend strongly on the dimensions and placement of the collimator aperture.

The collimator not only defines the effective field of view but also shields the detector from unwanted radiation. To effectively accomplish this latter purpose, the collimator material should also cover the rear of the detector. This is usually easy to achieve with portable NaI detectors but requires more effort when germanium detectors are used.

Intensive 50-100 keV X-ray radiation and 60-keV Am-241 gamma ray radiation are often emitted by process equipment, and this radiation can tie up the detector electronics unnecessarily. To alleviate this problem, a 1.5-mm-thick layer of lead (on the outside) and a 0.75-mm-thick layer of cadmium (on the inside) should be placed against the front face of the detector. This graded energy shield will absorb most of the low-energy photons incident on the detector without substantially reducing the number of gamma rays detected in the 375 to 450 keV range.

2.1.3 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. Either recalibrating one or more collection zones and comparing the results to previous analyses or testing the instrument with an appropriate check source is appropriate. When the response 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.

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 plutonium source (containing ~0.5 g plutonium) 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 to be adjacent to the crystal when the plug is in place.

The check source is fabricated in a manner to ensure its internal stability. Other than radiations increasing from the ingrowth of Am-241, the emission rate of the check source should remain constant.

2.2 Neutron Assay

Neutrons are emitted in the spontaneous fission of Pu-238, Pu-240, and Pu-242 and through the interaction of emitted α particles with certain light nuclei. These neutrons suffer little attenuation in passing through uranium or plutonium or through most structural and containment materials. Glovebox windows may reduce the energy of emerging neutrons, but, because of their regular and constant shape, their effect can generally be factored into the assay calibration.

To be useful for the assay of plutonium holdup, the neutron production rate per gram of plutonium must be known. The spontaneous fission contribution to the total neutron production can be computed from basic nuclear data after the isotopic composition of the contained plutonium has been determined. Computing the (α,n) contribution requires a knowledge of the chemical form of the plutonium and the amount and distribution of certain high-(α,n)-yield target materials.

The background count rate from neutron detectors may be a substantial part of the observed activity, often corresponding to as much as 20 g of plutonium in typical holdup assays. Thus, neutron assay is primarily applicable to the measurement of significant accumulations of plutonium.

The measured neutron yield from prepared calibration standards is used to calibrate each neutron assay collection zone. In the appendix to this guide, a method is given to calculate the anticipated neutron yield. This method provides the ability to calculate the neutron yield when the isotopic or impurity composition of the plutonium holdup is different from that of the calibration standards. The method can be used to calculate a ratio of the neutron production rate of the unknown material to the neutron production

rate of the standard material. The yield from the holdup material is then determined by multiplying the measured "known" material yield by the computed ratio.

2.2.1 Neutron Detection Instruments

To effectively employ the spontaneous neutron yield as a measure of plutonium holdup, it is necessary to detect the neutrons in the presence of a more intense gamma ray background to collimate the detector so that neutrons emanating from the collection zone under assay are preferentially detected.

Holdup assay is performed under in-plant conditions where ruggedness, portability, high neutron efficiency, and low gamma sensitivity in the detectors are important. He-3 has one advantage over BF_3 detector tubes in that the operating voltage for He-3 tubes does not increase as rapidly with increased gas pressure.

To increase the efficiency of the system, detector gas pressure in the tubes may be increased or multiple detectors may be connected in parallel to feed a common preamplifier.

He-3 and BF_3 detectors have efficiencies that increase as the energy of the neutrons decreases. To take advantage of this characteristic, the detectors may be surrounded by a neutron-moderating material such as polyethylene. The thickness of the moderator is important. When the moderating distance is short, a fraction of the higher energy neutrons passes through the gas chamber without being detected. Conversely, when the moderating distance is too long, a substantial number of low-energy neutrons are absorbed by the hydrogen contained in the moderator. A balance between these two effects is reached when the spacing between adjacent tubes is approximately 1 inch of polyethylene, and the spacing between the front of the unit and the detectors and the back of the unit and the detectors is approximately 1 3/4 inches when 1-inch-diameter tubes are used, and approximately 1 inch when 2-inch-diameter tubes are used.

To shield the detector from low-energy neutrons that may produce a complicated response pattern, the moderator material is covered with a thermal neutron absorber. Cadmium sheeting approximately 0.075 cm thick may be used for this application.

2.2.2 Collimators for Neutron Detectors

To assay a specific collection zone in the presence of other distributed sources of plutonium, it is necessary to collimate the detector. This is

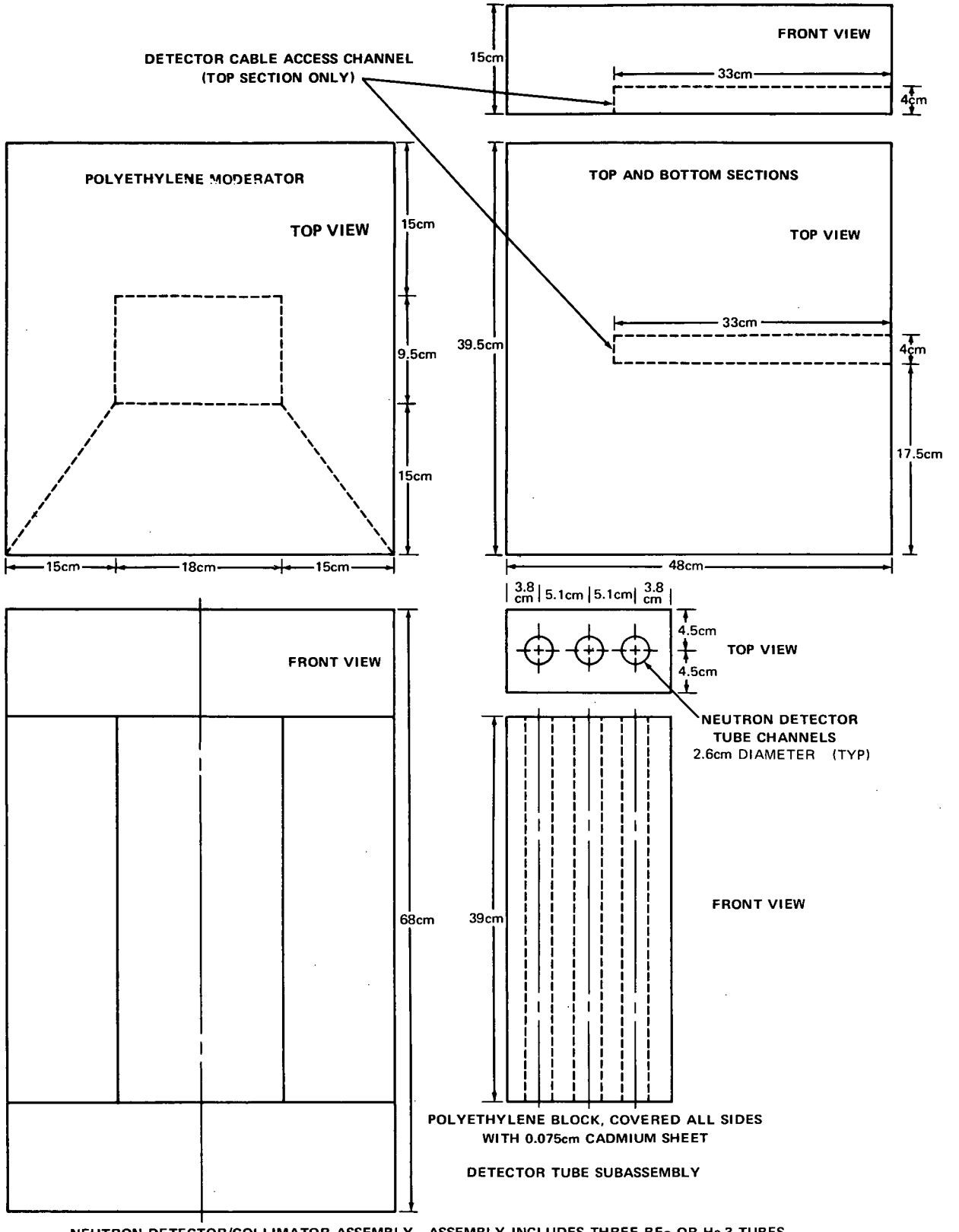
accomplished by stopping neutrons coming to the detector from all directions except the desired one. The cadmium surrounding the detector will stop essentially all neutrons striking the detector with energies below 0.4 eV. By adding moderator material around the outside of the detector in all directions except for the collimator channel, neutrons coming from unwanted directions will lose energy in this shield and will be absorbed in the cadmium cover. For each 6 inches of polyethylene added, the collimator assembly provides a factor of approximately ten in the directionality of the response.

An example of a collimated neutron detector assembly for plutonium holdup assay is shown in Figure 1. This assembly has a polyethylene shield thickness of 6 inches and 20 to 1 directionality. The combined weight of the detector and collimator exceeds the requirements for a hand-held probe. For this reason and to provide for reproducible positioning at each assay, a sturdy cart housing such a detector and its associated electronics is recommended. In order to assay items at different heights, it should also be possible to raise and lower the assembly to reproducible settings.

An example of a smaller, commercially available hand-held detector is given in References 5 and 6. This Shielded Neutron Assay Probe (SNAP) is 12 inches high and 10 inches in diameter and contains two He-3 detectors. It includes a 2-inch-thick polyethylene shield that provides 3 to 1 directionality. The SNAP has been used to measure plutonium holdup, UO_2F_2 holdup, and UF_6 enrichment. It is recommended for the assay of well-defined concentrations of plutonium in pumps, grinders, pipe elbows, or other items of equipment where portability and accessibility are more important than directionality.

A third example of holdup measurement by neutron detection is given in Reference 7. In this case, a completely uncollimated polyethylene slab containing a row of He-3 detectors was suspended in mid-air in some of the processing rooms of an industrial plutonium facility. The response of the detector was found to be proportional to the total room holdup if the plutonium was reasonably uniformly distributed and if the room was isolated from external sources. The calibration procedure for the use of this detector will not be described here. However, it is recommended as a means for quickly verifying total room holdup when measurements of individual items or equipment are not needed.

COLLIMATED NEUTRON DETECTOR ASSEMBLY FOR PLUTONIUM HOLDUP ASSAY



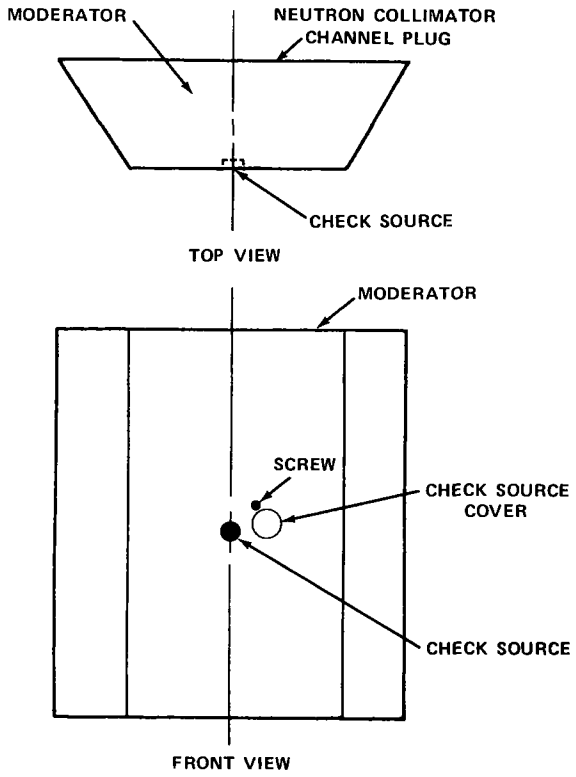
NEUTRON DETECTOR/COLLIMATOR ASSEMBLY. ASSEMBLY INCLUDES THREE BF_3 OR He-3 TUBES (2.54cm DIAMETER) UNIT CAN BE MODIFIED TO INCREASE OR DECREASE THE NUMBER OF TUBES. MODERATOR THICKNESS OF 15cm PROVIDES ~10:1 DIRECTIONALITY. ADDITIONAL POLYETHYLENE CAN BE ADDED TO IMPROVE DIRECTIONALITY (e.g., 30cm POLYETHYLENE PROVIDES ~100:1 DIRECTIONALITY). COMPONENTS ARE BOLTED OR STRAPPED TO REMAIN IN A FIXED CONFIGURATION.

Figure 1

2.2.3 Check Source for Neutron Assay

To ensure the proper operation of the neutron assay system prior to making an assay, it is necessary to test the response of the instrument. An appropriate neutron assay check source can be measured, or the detector response for one or more collection zones can be recalibrated and compared to the results of previous calibrations.

An appropriate neutron assay check source can be prepared by implanting a small encapsulated plutonium source (containing about 5 g of plutonium) into the face of a plug of neutron moderating material (see Figure 2). The plug is fabricated to fit and close the collimator channel. When the response from the check source remains within the expected value, the previous calibration data are assumed to be valid.



NEUTRON COLLIMATOR CHANNEL PLUG AND CHECK SOURCE

Figure 2

2.3 Thermoluminescent Dosimeter (TLD) Assay

Crystals of LiF, CaF_2, $CaSO_4$, or other compounds can store energy at manganese or dysprosium impurity centers when they are struck by gamma or neutron radiation. At some later time, the crystals can be heated rapidly to several hundred °C to induce "thermoluminescence." The light output at this time is proportional to the amount of radiation received. Thermoluminescent detectors that are primarily gamma sensitive, graded X-ray shields, read-out instrumentation, and other accessories are commercially available.

TLDs have been used to measure the holdup in gloveboxes by placing them at regular intervals on the outside surfaces. The TLDs are left in place overnight in order to accumulate a measurable dose. Accuracies of ±20 percent relative to cleanout values are reported for plutonium of known isotopic composition. TLDs have also been used to measure the holdup in the interior of large furnaces that are not accessible by other means. For both of these examples, calibration requires either careful dose and geometry calculations or mockups of the actual collection zone. Because their use is relatively new and only a few published references exist (Refs. 8 and 9), TLDs will not be discussed further in this guide. However, they should be kept in mind for special applications.

3. ISOLATION OF COLLECTION ZONES

To ensure that each collection zone is independently assayed, it is necessary to shield the detector from all radiations except those radiations emanating from the collection zone being assayed. This is principally accomplished through the use of the collimators described in Sections B.2.1.2 and B.2.2.2. Two additional means exist to isolate a collection zone.

3.1 Detector Positioning

An unobstructed side view of a collection zone is preferred. When plutonium is located behind the zone under assay in another collection zone or a storage facility, either consider performing an additional background assay with the detector above or below the collection zone and pointing at the material behind the zone under assay, or consider the use of shadow shielding.

3.2 Shadow Shielding

It may not be possible to avoid interfering radiations through the collimator design or through choosing the detector position for assay. In such cases, 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 very thick and its dimensions match or exceed the back side of the collection zone under assay, no interfering radiations will penetrate through the shadow shield to the detector. While such characteristics are desirable, the size of such a shield would limit its transportability. A rectangular panel mounted on wheels as an upright panel and containing ~5 cm of neutron moderator (e.g., benelex, WEP, or polyethylene) and ~0.5-cm lead sheet is recommended. To use such a panel, it is necessary to measure the response of the collection zone with and without the shield in place. Also, the gamma and/or neutron transmission factor of the shield itself should be measured beforehand with a representative plutonium sample. From these measurements, the assay of the collection zone can be corrected for background radiation transmitted through the shield.

4. CALIBRATION FOR HOLDUP MEASUREMENTS

4.1 Basic Counting Geometries

There are three fundamental counting geometries that can be used to represent most collection zones. 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 that is 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 deposits

of plutonium could exhibit very large gamma ray self-attenuation and could therefore require great care in analysis or could require neutron assay.]

4.1.2 Line Source

If the material being assayed is distributed along a linear path so that only a segment of that distribution length is contained in the detector field of view, the zone 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 ducts or columns.

4.1.3 Area Source

If the material being assayed is spread over an area large enough so that the material covers the full field of view of the detector for a range of source-to-detector distances, the zone 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; so the source-to-detector distance can affect the instrument response and should 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 large rectangular ducting.

4.2 Calibration of Detector Response

4.2.1 Mockup of Known Material Distributions

When a gamma ray assay is used and a collimator setting has been selected, the detector response for the three basic source distribution geometries listed above should be determined. For the point source, the response is expressed as "counts/min/gram Pu-239" at a specified source-to-detector distance. For the line source, the response is expressed as "counts/min/(gram Pu-239 per unit length)" at a specified source-to-detector distance. For the area source, the response is expressed as "counts/min/(gram Pu-239 per unit area)" at a specified source-to-detector distance. When

neutron assay is used, the response for a point source is expressed as "counts/min/(gram of Pu-240 effective)" at a specified source-to-detector distance. Calculation of Pu-240 effective from the plutonium isotopic composition is described in the appendix to this guide. Analogous expressions can be given for line and area sources although neutron assay is usually restricted to dense, isolated items of equipment that can be represented as point sources. For both neutron and gamma measurements, corrections to the point and line source calibrations for different detector distances are made using the $1/r^2$ or $1/r$ count-rate dependence, respectively. For further detailed discussion of the measurement of detector responses for these basic geometries, see Reference 10.

For gamma ray assay, the calibration of the point source response can be accomplished with a well-characterized encapsulated standard plutonium 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.* Care must be taken in the preparation of this calibration standard to ensure that the amount of encapsulated Pu-239 is well known. It is also important to measure the gamma ray attenuation through the encapsulating material and the self-attenuation of the plutonium foil and to correct the calibration standard response to compensate for these effects. The amount of Pu-239 encapsulated in this standard should provide sufficiently strong count rates for good statistical precision of the calibration in a reasonable period of time.

For neutron assay, it is probably necessary to encapsulate a larger amount of material in the calibration standard because the spontaneous neutron production rate is significantly less than the 375 to 450 keV gamma ray production rate. A quantity of 50 to 100 g of plutonium should be adequate for most applications. Again it is important to know the exact quantity and isotopic composition of the plutonium. Also, the neutron calibration standard may generate more neutrons than directly attributable to the spontaneous fission and (α,n) reactions. Because a relatively large quantity of PuO_2 is encapsulated in the neutron assay calibration standard, some spontaneous

*It should be noted that a calibration source may be used as a check source, but a check source should not be used as a calibration source.

fission or (α,n) neutrons may be absorbed in Pu-239 or Pu-241 nuclei, producing additional neutrons through the induced fission reaction. The amount of multiplication depends in a complex manner on the amount and distribution of PuO_2 and on the surrounding medium (Ref. 11). For 50 g distributed in the bottom of a 4-inch-diameter can, a self-multiplication of 0.5 percent of the total neutron output would be typical. At 100 g, 1 to 2 percent may be expected. Thus, this effect is typically smaller than other errors associated with holdup measurements and can be neglected if the standard contains 100 g or less of well-distributed material. The chemical and isotopic composition of the plutonium will have a larger effect, as described in the appendix to this guide. The measurement of the line source response is best accomplished by constructing a cylindrical surface distribution of plutonium with the aid of large foils. It is also possible to establish the line source response using a point source, as described in Reference 4. The line source geometry is closest to that of the pipes and ducts likely to be encountered in actual measurements.

The area source response can be measured with the same plutonium foils, laid flat to simulate the expected distribution on surfaces such as walls and floors. The area response can also be established using a point source. The point source is measured at different radial distances from the center of the field of view of the collimated detector. The response at each radial distance is weighted by the area of a concentric ring at that radius. From these weighted responses, it is then possible to calculate the area of a circular region of uniform plutonium deposition that would yield the same total response as the point source. From this equivalent area, the expected response/(gram Pu-239 per unit area) can be derived. Further useful details on this procedure may be found in Reference 12. For both line and area calibrations, the self-attenuation of the foils or point sources should also be taken into account.

There may also be special material distribution geometries that are known to exist in the facility but are not readily represented by one of the three basic configurations described above. These special geometries should be mocked up, as carefully as possible, using the large plutonium foils and/or 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 Ref. 10).

Because material particle sizes (or material deposit thicknesses) have a significant effect on the self-attenuation of the gamma ray signals, one should use (whenever practical) well-characterized process material for preparing calibration standards and should duplicate to the extent possible process holdup distribution relative to particle size and/or thickness. Furthermore, holdup in floors is often deposited at varying depths into the floor, rather than on the surface. Thus, calibration standards for such measurements should 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. Whenever possible, holdup measurements before and after the cleanout of a piece of shutdown process equipment should be taken to establish this comparison history and to improve the accuracy of the calibration for each collection zone.

4.2.2 Measurement of Calibration 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 should be aware of occasions when new equipment is brought into the plant for installation in the process. At that time, before installation, calibration sources can be conveniently 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 LIMITS OF ERROR

The measurement of holdup in a complex plant environment can involve a very large number of measurements, as is implied in the text to follow. 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:

a. Careful and extensive holdup measurements are made infrequently (e.g., annually) and
b. At more frequent intervals (e.g., at inventory times), careful measurements are again made in the (presumably fewer) known problem areas, and "spot check" measurements are made in the other, 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.

5.1 Holdup Measurements

In performing the holdup measurements, one must be aware of the large variability in holdup assays arising primarily from variability in the measurement conditions (e.g., background, geometry, gamma ray and/or neutron attenuation, material distribution). Accordingly, every effort should be made to perform the assays from as many vantage points as possible for each collection zone. Where this may be impractical on a routine basis because of time or space constraints, one might consider multiple measurements initially on a collection zone, followed by fewer routine measurements at representative assay sites. Careful thought in the selection of measurement points and measurement strategy will minimize ambiguities in the interpretation of the data.

5.1.1 Selection of Collection Zones and Detector Positions

Location and configuration of collection zones should be established on the basis of a detailed physical examination and a radiation survey of the physical layout of the facility. Preliminary measurements should be made 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 the collection zone. More than one detector position may be required.

In the cases where radiation surveys have pointed out zones of high holdup collection, extra care will be necessary in the holdup measurements for those zones to minimize their contribution to the overall holdup variability. [Where radiation surveys show little holdup, proportionately less time should be budgeted.] Selection of optimum detector positions should also include 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 Measurement Procedure

The measurement and analysis of gamma or neutron radiation from a collection zone should be carried out by treating the material distribution as a point, line, or area source, as 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 reveal 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, these positions should be permanently marked to ensure reproducibility of the subsequent measurements. The markings should indicate detector location (including height) and orientation. Each assay site should also be uniquely labeled to facilitate unambiguous reference to each measurement and its location in the assay log. Furthermore, assay site labels and markings should indicate whether neutron or gamma ray measurements are to be made. Alphabetic labels (for example, "G" for gamma and "N" for neutron) and color-coded tape markings of the sites would be useful. The markings should be protected (for example, with clear epoxy) to ensure their long-term durability.

At each detector position in a given collection zone, the gamma or neutron radiation intensity should be measured. The line-of-sight background should then be measured by moving the detector and collimator to one side (still pointing in the same direction as during the assay) and measuring the radiation intensity from the surrounding materials. During the background measurement, the vessel in which the holdup is being measured should not be in the field of view of the detector. Because uncertainties in geometry, attenuation, or sample matrix will usually dominate the total response variability,

the counting period need not be long. Having 1000 to 10,000 net counts is generally sufficient for most holdup applications.

This procedure should be repeated at all measurement positions and in all counting geometries designated for each collection zone. The final holdup value for the zone should be 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 should be assayed in a variety of ways. For example, one could measure an apparatus up close and treat it as an area source; the measurement could then be repeated at a large distance, 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 an indication 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 375 to 450 keV gamma rays, it is necessary to correct each assay for attenuation of the signal, either within the plutonium holdup material or by structural materials. Without this critical correction, the assay is no more than a lower limit on the true holdup value. The attenuation correction may be based on calculations of known attenuation in uniform materials, on earlier measurements of materials similar to those found in the plant equipment, or on direct measurements of gamma ray transmission through the actual equipment. Details on establishing an appropriate attenuation correction are given in Laboratory Exercise #4 of Reference 4. Additional treatment of gamma ray attenuation corrections is given in Reference 13.

5.1.4 Gamma Ray Interferences

Variability in the observed gamma ray response may arise as a result of the presence of extraneous gamma ray emitters or as a result of fluctuations in the background from the Compton scattering of higher-energy gamma rays. The magnitude of this effect is generally small. It can be monitored by

observing the spectrum with a multichannel analyzer, but, unless data on periodically recovered holdup accumulations are in error, this contribution can be ignored.

5.1.5 Matrix Effects on Neutron Assay

A change in the neutron yield for a plutonium sample of fixed isotopic content can be caused by a change in the concentration of high-(α,n)-yield impurities in the matrix. If it is possible to estimate the range of permissible impurity concentrations, the variation in a typical neutron yield can be calculated using the method given in the appendix to this guide.

5.1.6 Effect of Isotopic Uncertainty

Gamma ray measurements of plutonium holdup provide a direct determination of the fissile plutonium (i.e., Pu-239 and Pu-241) holdup in the zone under consideration. On the other hand, neutron techniques measure only the Pu-240 effective content, and chemical techniques provide elemental analysis without consideration of the isotopic makeup. Thus, knowledge of the isotopic composition of the plutonium is necessary to correlate holdup measurements with chemistry and accountability values. Gamma ray assays must be divided by the Pu-239 isotopic fraction, and neutron assays must be converted from Pu-240 effective to total plutonium in order to express holdup in terms of total plutonium.

If the process equipment is thoroughly cleaned each time the isotopic composition is changed, the holdup may consist primarily of the current material. In that case, the declared isotopic composition can be used. When mixing occurs, use of the stream-averaged isotopic composition is appropriate. Bounds on the isotopic composition are estimated by considering the batches of highest and lowest composition and computing the corresponding range. This measure of variability must then be incorporated into the estimated holdup standard deviation before making direct comparisons with the chemical analyses. In general, gamma ray measurements of Pu-239 will be less sensitive to isotopic variations than neutron measurement of Pu-240.

5.2 Assignment of Limits of Error

The assignment of a limit of error (i.e., twice the standard deviation) to a holdup measurement is extremely difficult on a rigid statistical basis.

This is because the only statistically predictable fluctuations (e.g., counting statistics) in this application are frequently negligible in comparison with variability due to counting geometry (including material distribution), gamma ray attenuation, gamma ray background and interferences, neutron matrix effects, and instrument instabilities. One should therefore recognize that the variability can be large and guard against underestimating the standard deviation of the overall holdup value in a collection zone. Careful measurements should be carried out 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 10.

A reasonable estimate of the standard deviation of the measured holdup for a given collection zone may be obtained by consideration of 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. The standard deviation (σ) for that mean value is estimated conservatively as one-half the range of holdup values obtained in the measurements.

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

$$\sigma_{(h-u)} = \sigma_{(stat)} + \sigma_{(meas)} \qquad (1)$$

5.3 Estimation of Bias

When a single collection zone is cleared out, where possible a holdup assay should be performed before (H_{before}) and after (H_{after}) the cleanout. By comparing the amount of plutonium removed (Pu_r) to the recovery amount predicted through the in situ holdup assays (Pu_a), the collection zone calibration can be updated, and the calibration and assay standard deviations can be

based on relevant data. The amount of plutonium recovered (Pu_r) during the cleanout of a specific collection zone can be assayed through sampling and chemical analysis, through calorimetry, or through other applicable non-destructive assay methods (e.g., spontaneous fission coincidence detection or gamma ray assay).

The assay value for the recovered amount is computed as the difference in the holdup assays before and after the cleanout:

$$Pu_a = H_{before} - H_{after} \qquad (2)$$

The percent difference (Δ) between the assay and recovery values for the plutonium holdup,

$$\Delta = (Pu_a - Pu_r)/Pu_r \quad , \qquad (3)$$

is then computed. A running tabulation of the quantities Pu_a, Pu_r, and Δ (as well as their standard deviations, σ_a, σ_r, and σ_Δ) should be kept in the assay log for each collection zone.

The average value ($\bar{\Delta}$) of the percent differences between Pu_a and Pu_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_Δ) of the percent differences (Δ_i) from their mean value ($\bar{\Delta}$) serve as a check on the appropriateness of the size of the estimated standard deviation of the holdup measurements. To the extent that the standard deviation of Pu_r is small compared with the uncertainty in Pu_a (usually an adequate assumption), the quantity s_Δ should be comparable in size to the standard deviation of Pu_a. For K measurements of the percent differences Δ_i for a given collection zone, the quantity s_Δ is given by:

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

Equation (4) assumes that all the σ_Δ's are equal. For a calculation of s_Δ using weighted sums, see Reference 14.

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

C. REGULATORY POSITION

To develop a program for the periodic in situ assay of plutonium residual holdup as an acceptable measurement method for this inventory component, it is necessary to consider the factors in the following sections.

Care must be exercised during the fabrication and use of check sources and calibration standards to ensure their continued integrity and to prevent contamination. In addition, the usual precautions for safeguarding plutonium should be taken.

1. DELINEATION OF COLLECTION ZONES AND ASSAY SITES

Preliminary radiation survey measurements of the plutonium processing facility should be used to budget the measurement time to emphasize high-holdup areas, to establish independent collection zones, and to determine detector positions within the zones.

a. 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 an 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.

b. Each assay site should be permanently marked with paint or colored tape on the floor to ensure reproducible assay positions. The markings should be protected (for example, with clear epoxy) to ensure their long-term

durability. Detector height and orientation should be clearly indicated in the assay log for each measurement site and, if possible, included with the site markings.

c. Each assay site should be uniquely labeled to facilitate unambiguous reference to that site in the assay log. A labeling and tape color coding convention should be established to distinguish neutron assay sites from gamma ray assay sites.

d. Gamma ray assay should be used for collection zones containing less plutonium than the neutron detection limit. Also, gamma ray assay should be used for all structures that do not contain irregularly shaped components capable of significantly attenuating the emerging gamma rays. Neutron assay should be used for all structures not suitable for gamma ray assay. There may be some large structures such as furnaces that can be measured only with small interior probes or with thermoluminescent dosimeters.

e. Areas may be denoted as "problem areas" so that careful holdup measurements will be made in these areas each time plant holdup is to be determined; or the 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 INSTRUMENTS

Neutron and gamma ray assay capability can be provided if desired using separate or compatible electronics with interchangeable detector probes. Compatible electronics can provide for both He-3 or BF_3 neutron detection and NaI(Tl) gamma ray detection. The electronics unit should have a temperature coefficient of less than 0.1 percent per °C. Battery-powered electronics can expedite assays.

2.1 Gamma Ray Assay

Gamma ray assay should be based on the activity observed in the energy range from 375 keV to 450 keV, excluding the composite gamma ray complex centered at 333 keV. Yield data for appropriate gamma rays are presented in Section B.2.1 of this guide.

2.1.1 Detector Selection

Gamma ray detectors for holdup measurements should have FWHM (full width at half maximum) resolution better than 10 percent at 662 keV (Cs-137 gamma ray). NaI(Tl) detectors can exhibit resolutions as good as 7 percent and are suitable for this application. The crystal depth should be sufficient to detect a significant percentage of 400-keV gamma rays. For NaI(Tl), the minimum depth should be 1 inch. A 2-inch depth is recommended.

The crystal should be stabilized with a suitable radioactive source. An internal seed containing Am-241 is recommended for this application. The electronics should be capable of stabilizing on the reference radiation emitted by the seed. The crystal face (external to the cover) should be covered with 0.075 mm of cadmium and 1.5 mm of lead to filter low-energy radiations.

Two single-channel analyzers should be provided with lock-set energy windows. One channel should be set to admit gamma rays from 375 to 450 keV. Unless equilibrium of the U-237 and Pu-241 can be ensured, the 333-keV region of the gamma ray spectrum should be completely excluded. The second channel should be set above the first window to provide a background subtraction for the assay window. This second window should be set from approximately 475 to 575 keV. The width and position of this window is a matter of personal preference in how the background subtraction should be done. These analyzers should be integrally packaged, get their signals from the same detector, and process signals with the same electronics.

2.1.2 Gamma Ray Collimator

A cylinder of shielding material such as lead should be made concentric 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 facility (1.5 cm of lead thickness should be sufficient for most applications). The collimator sleeve should be fixed over the end of the detector crystal at a reproducible setting identical to that used in the calibration measurements.

2.1.3 Gamma Ray Calibration and Check Sources

Standard sources of Pu-239 should be provided for calibration of the measurement system for the basic measurement geometries described in Section B.4. A small encapsulated plutonium sample can be used both as a

calibration standard for the point source counting geometry and as a check source for verification of instrument stability. For the line and area calibrations, large plutonium foils can be used, or the calibrations can be derived from a series of measurements made with the point source. The gamma ray self-attenuation correction should be clearly specified for all foils and samples.

2.2 Neutron Assay

2.2.1 Neutron Detector Selection

Neutron detectors should have high detection efficiency and be capable of operating in the presence of gamma radiation. He-3 and BF_3 neutron detectors are recommended for this application. Multiple detector tubes with matched operating performance should be connected in parallel to a single preamplifier to increase the overall detection efficiency. Neutron detectors should be surrounded by a layer of neutron moderator material to enhance their detection efficiency. The neutron moderator layer should be covered with a low-energy neutron absorber to filter out extraneous neutrons from the desired signal.

2.2.2 Neutron Collimator

A slab collimator or concentric cylinder collimator of polyethylene should completely surround the detector, leaving open only a detection channel in one direction. The moderator thickness should be selected to provide the directionality required for each facility. A directionality profile providing a 10:1 response ratio is desirable. However, for portable detectors a 3:1 ratio may be used.

2.2.3 Neutron Calibration and Check Source

A 50 to 100 g sample of plutonium should be adequate both as a point source calibration standard and as a check source.* The isotopic composition, Am-241 content, and high-(α,n)-yield impurity composition should be representative of the plutonium being processed. The neutron yield of the standard should be independently measured, if possible, and also computed using the method described in the appendix of this guide. If the measured and calculated yields differ by more than 20 percent, any future yield calculations should be normalized to be consistent with this measurement.

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*Although a well-characterized calibration source may be used as a check source, a check source should never be used as a calibration source.

2.3 Service Cart

A cart carrying electronics and both detector probes should be provided. The capability to raise or lower the probes to reproducible settings should be included.

3. CALIBRATION

3.1 Instrument Check

The stability of the neutron and gamma ray detection systems should be tested prior to each inventory by comparing the observed counts obtained from the check source, minus the counts with the shaped shield in place but without the check source, to the readings obtained prior to previous inventories. If the 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), all previously established calibrations using this detection system should be considered valid. If the measurement is not consistent, the operation of the unit should be checked against the manufacturer's recommendations and repaired or recalibrated, as required. These check source measurements should be supplemented with regular remeasurements of instrument calibrations to ensure continued proper instrument performance over the entire operating range.

3.2 System Response Calibration

The response of the detection system should be determined with well-known quantities of plutonium 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 should have a separate section in the assay log, with the corresponding calibration derived provided 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 plutonium from the calibration in addition to position and instrument electronic setting verification. There should also be provision 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 isotopic composition of the plutonium processed during the current operational period should be determined. Variations in the neutron and 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 difference.

Prior to each inventory, the operation of the neutron and 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 plutonium 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 the floor location, probe height, and probe orientation. The electronic settings should be verified every 1 or 2 hours with the check source. During the actual assay of the collection zones, the check source should be removed or shielded so as not to interfere 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 plutonium remain within the collection zone. The operator should initial the measurement log to ensure compliance for each collection zone.

When the preceding steps have been completed, the measurement at each collection zone should be taken, recorded, and converted to grams plutonium. If each value is within an expected or permissible range, the assayist can proceed to the next collection zone. However, if the collection zone contains an unexpectedly large amount of plutonium, it 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.

5. ESTIMATION OF HOLDUP ERROR

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 systematic errors 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 any case, all calibrations should be updated at least once per year.

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APPENDIX

A. Neutron Yield Computations

The following model for the calculation of the total spontaneous neutron yield from plutonium-bearing materials assumes that the plutonium is widely dispersed. With this condition, there will be no significant neutron production through induced fission of Pu-239 or Pu-241. The total neutron yield of plutonium holdup will then be the sum of the spontaneous fission and (α,n) contributions:

$$Y_n = Y_{SF} + Y(\alpha,n) \tag{1}$$

1. SPONTANEOUS FISSION NEUTRONS

To determine the spontaneous neutron yield of plutonium, the isotopic composition must be known. (The contribution from U-238 spontaneous fission is usually negligible even if uranium is present in large quantities.) The yield from the plutonium isotopes is given by:

$$Y_{SF} = M_{238}\, Q_{238} + M_{240}\, Q_{240} + M_{242}\, Q_{242} \tag{2}$$

where M_i is the total mass of the ith plutonium isotope, and Q_i is the spontaneous fission neutron yield per gram of the ith isotope. Using the yield data from Table A-1, Equation (2) can be rewritten as:

$$Y_{SF} = (1030 \text{ n/sec-gram})\, M_{240}(\text{effective}) \tag{3}$$

where
$$M_{240}(\text{effective}) = 2.50\, M_{238} + M_{240} + 1.70\, M_{242} \tag{4}$$

The coefficients 2.50 and 1.70 are the spontaneous fission yields of Pu-238 and Pu-242 relative to Pu-240. The concept of effective Pu-240 mass reflects the fact that most of the spontaneous fission yield is due to that isotope.

Table A-1

ALPHA-PARTICLE AND SPONTANEOUS FISSION NEUTRON YIELDS

Nuclide	Half-life (yr)	Alpha Activity (α/sec-gram)	Q_i Spontaneous Fission (n/sec-gram)	Y_i $PuO_2(\alpha,n)$ Yield* (n/sec-gram - Pu)
Pu-238	87.78	6.33×10^{11}	2.57×10^{3}	1.4×10^{4}
Pu-239	24,150	2.30×10^{9}	2.22×10^{-2}	42.5
Pu-240	6,529	8.43×10^{9}	1.03×10^{3}	157
Pu-241	14.35**	9.39×10^{7}	2.43×10^{2}	1.3
Pu-242	379,000	1.44×10^{8}	1.75×10^{3}	2.2
Am-241	433.8	1.27×10^{11}	6.05×10^{-1}	2957
U-234	2.47×10^{5}	2.29×10^{8}	5.67×10^{-3}	4.65
U-235	7.1×10^{8}	7.93×10^{4}	5.96×10^{-4}	1.37×10^{-3}
U-238	4.51×10^{9}	1.23×10^{4}	1.12×10^{-2}	1.93×10^{-4}

*Oxygen yield from PuO_2 form only.
**α-branching ratio: 2.46×10^{-5}.

2. (α,n) NEUTRONS

When the plutonium holdup is in the form of oxide, the major contribution from (α,n) reactions will be due to the O-18(α,n) Ne-21 reaction. The additional neutron yield is typically 50-100 percent of the spontaneous fission yield. The (α,n) yield can be calculated from the yields per gram of each isotope of Pu(Y_i) given in Table A-1:

$$Y(\alpha,n)\ \text{oxide} = \sum_i M_i Y_i \qquad (5)$$

The summation over M_i should also include Am-241, which is a strong alpha emitter.

In addition to (α,n) production in the oxide itself, certain low-Z impurities in the oxide can contribute substantially. Values for the yields of neutrons obtained in bombarding thick targets of these elements with 5.2-MeV alpha particles are given in Table A-2.* Further research may change these values somewhat, but they are sufficient for computing the approximate effect of these elements if they exist as impurities in PuO_2. One method for doing this is to compute the impurity (α,n) yield relative to the oxide (α,n) yield.

$$Y(\alpha,n)\ \text{impurity} = Y(\alpha,n)_{\text{oxide}} \sum_j (P_j A_o I_j)/(P_o A_j I_o) \qquad (6)$$

P_j is the (α,n) neutron yield in the impurity element, and P_o is the yield in oxygen (0.050 neutrons/10^6 alphas). A_j is the atomic weight of the impurity element, and A_o = 16 for oxygen. I_j is the impurity concentration expressed in parts per million (by weight) of plutonium oxide. I_o is the concentration of oxide expressed in the same way, 118,000 ppm. If the impurity concentration is expressed as ppm of plutonium, it can be converted to ppm of plutonium oxide by multiplying by the gravimetric dilution factor, 0.882.

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*Most of the yields in Table A-2 are based on: J. K. Bair and J. Gomez del Campo, "Neutron Yields from Alpha-Particle Bombardment," Nuclear Science and Engineering, Vol. 71, p. 18, 1979.

Table A-2

(α,n) NEUTRON YIELDS FOR VARIOUS LIGHT ELEMENTS

Element	Neutron Yield per 10^6 alphas = P_j
Be	58
B	18
F	6.4
^{7}Li	1.3
Na	≃1.5
Nat_{Mg}	0.89
Al	0.44
Nat_{Si}	0.077
Cl	≃0.05
C	0.05
O	0.050
N	0.00
As	0.00

*Most of these yields are based on: J. K. Bair and J. Gomez del Campo, "Neutron Yields from Alpha-Particle Bombardment," Nuclear Science and Engineering, Vol. 71, p. 18, 1979.

To summarize the calculation of (α,n) neutron yields in oxide that also contains impurities, $Y(\alpha,n)$ from all sources is given by:

$$Y(\alpha,n) = \sum_i M_i Y_i \left[1 + .0027 \sum_j P_j I_j / A_j\right] \qquad (7)$$

Elements other than those listed in Table A-2 yield no neutrons by (α,n) reactions for the alpha energies obtained from plutonium and americium decay. Also note that the summation over i must include Am-241 and that the summation over j includes only the oxygen that is not bound up as plutonium oxide.

3. SAMPLE CALCULATION FOR PuO_2 - UO_2

Consider the case of 1 gram of recycle plutonium blended to 3 percent by weight PuO_2 in a UO_2 matrix where the isotopic composition is as given in Table A-3. For mixed oxides, the oxygen density is approximately the same as in PuO_2 alone. Also, plutonium and uranium have similar atomic numbers. For these reasons, it may be assumed that the oxygen (α,n) yield in mixed oxide is the yield in PuO_2, further reduced by the blending ratio, $PuO_2/(PuO_2 + UO_2)$.

Using the isotopic composition given in Table A-3 and using Equation (3), the spontaneous fission neutron yield can be found to be 218 n/sec for 1 gram of plutonium. Then the neutron production in the oxide can be calculated using the masses M_i of each isotope and the yields Y_i from the fifth column of Table A-1. The result of 112 n/sec is given in the last column of Table A-3. Note that the α-particle yield of plutonium is nearly constant in time, but that, because Am-241 builds up in time, the total alpha production increases at a rate of roughly 0.3 percent per month in typical reactor fuel.

The impurity (α,n) yields are calculated in Table A-4. The calculation is based on impurities in PuO_2 only. The mixed oxides are assumed to consist of blended PuO_2 and UO_2 particles approximately 40 μm in diameter where most alpha particles stop within the PuO_2 particles. If the particle size were smaller or the mixed oxide were created through co-precipitation, the uranium impurity content would also contribute to the plutonium (α,n) yield. In the present example, it is sufficient to use the neutron yields P_j from Table A-2, the concentrations I_j from Table A-4, and Equation (6) or (7).

Table A-3

SAMPLE CALCULATION FOR 1 GRAM OF PLUTONIUM

Nuclide	Isotopic Composition	Y_{SF} (n/sec)	Y(α,n) oxide (n/sec)
Pu-238	.003	8	42
Pu-239	.756	0	32
Pu-240	.185	191	29
Pu-241	.045	0	0
Pu-242	.011	19	0
Am-241	.003	0	9
Total Yields		218 (n/sec)	112 (n/sec)

Table A-4

IMPURITY (α,n) YIELD

Impurity	Arbitrary Concentration I_j in PuO_2 (ppm by wgt)	Impurity (α,n) Yield (112n/sec) (.0027) $P_j I_j / A_j$
Li	9	1
Be	8	16
B	10	5
C	200	0
F	125	13
O (moisture)	4600	4
Na	120	8
Total		47 (n/sec)

The total neutron yield from 1 gram of plutonium in PuO_2 is then 218 + 112 + 47 = 377 n/sec. Using the gravimetric dilution factor of .882, this is 333 n/sec for 1 gram of PuO_2. If the PuO_2 is blended so that $PuO_2/(PuO_2 + UO_2) = .03$, the neutron yield from 1 gram of mixed oxide is 10 n/sec.

The impurity (α,n) yields, P_j, used in this example are currently known to about 10 percent accuracy for most elements and 50 percent accuracy for the others. The oxide (α,n) yields, Y_j, are known to 10 percent or better. Both yield calculations must assume perfect mixing, however. For these reasons, neutron yield calculations are accurate to 10 percent at best, and the neutron holdup measurement calibration should be based on representative standards rather than calculation wherever possible.

B. Conversion of Measured M_{240}(effective) to Total Plutonium

To convert a measured effective Pu-240 mass to actual total plutonium, one must use both the relationship between these two quantities, as shown in Equation (4), and the known isotopic composition of the samples being measured. Let f_{238}, f_{239}, f_{240}, f_{241}, and f_{242} represent the weight fractions of the respective plutonium isotopes in the unknown sample. The Pu-240 effective weight fraction, f_{240}(effective), can be defined as:

$$f_{240}(\text{effective}) = M_{240}(\text{effective})/M_{Pu}\ (\text{total}) \qquad (8)$$

Then, use of Equation (4) and the definition of the individual isotopic weight fractions, f, gives:

$$M_{Pu}(\text{total}) = M_{240}(\text{effective})/f_{240}(\text{effective}) \qquad (9)$$

where

$$f_{240}(\text{effective}) = 2.50\ f_{239} + f_{240} + 1.70\ f_{242} \qquad (10)$$

Generally, as previously mentioned in this guide, the relative measurement uncertainty of M_{240}(effective) in a holdup measurement will be much larger

than that of f_{240}(effective), so the relative error in M_{Pu}(total) is essentially equal to that of M_{240}(effective).

As an example calculation, the sample of isotopic composition given in Table A-3 has an effective fraction given by:

$$f_{240}(\text{effective}) = 2.50(0.003) + 0.185 + 1.70(0.011)$$

$$= 0.211$$

Thus, a holdup measurement of 35 ± 10 grams Pu-240 effective corresponds to 166 ± 47 grams total plutonium, where the relative error in the total plutonium result was taken to be equal to that of the M_{240}(effective) result.

DRAFT VALUE/IMPACT STATEMENT

1. PROPOSED ACTION

1.1 Description

Licensees authorized to possess at any time more than 1 kilogram of plutonium are required by Part 70, "Domestic Licensing of Special Nuclear Material," of Title 10 of the Code of Federal Regulations to calculate a material balance based on a measured physical inventory at intervals not to exceed 2 months. 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 detailed in Regulatory Guide 5.13, "Conduct of Nuclear Material Physical Inventories."

Plutonium residual holdup is defined as the plutonium inventory component remaining in and about process equipment and handling areas after those collection areas have been prepared for inventory. This regulatory guide describes procedures acceptable to the NRC staff for the in situ assay of the residual plutonium holdup.

1.2 Need for Proposed Action

Regulatory Guide 5.23 was published in 1974. The proposed action, a proposed revision to this guide, is needed to bring the guide up to date with respect to advances in measurement methods, as well as changes in terminology.

1.3 Value/Impact of Proposed Action

1.3.1 NRC Operations

The regulatory positions will be brought up to date.

1.3.2 Other Government Agencies

Not applicable.

1.3.3 Industry

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

1.3.4 Public

No adverse impact on the public can be foreseen.

1.4 Decision on Proposed Action

The regulatory guide should be revised to reflect the improvement in measurement techniques and to bring the language of the guide into conformity with current usage.

2. TECHNICAL APPROACH

Not applicable.

3. PROCEDURAL APPROACH

3.1 Procedural Alternatives

Potential RES procedures that may be used for the proposed action include the following:

- Regulation
- Revision of regulatory guide
- ANSI standard, endorsed by regulatory guide
- Branch position
- NUREG report

3.2 Value/Impact of Procedural Alternatives

Since a usable and useful regulatory guide already exists and modifications are minimal, the procedure having the least impact is to revise the guide.

3.3 Decision on Procedural Approach

A revised regulatory guide should be prepared.

4. STATUTORY CONSIDERATIONS

4.1 NRC Authority

The authority for the proposed action would be derived from the Atomic Energy Act of 1954, as amended, and is implemented through the Commission's regulations, in particular, 10 CFR Part 70.

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

Regulatory Guide 5.23 should be updated.

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D.C. 20555