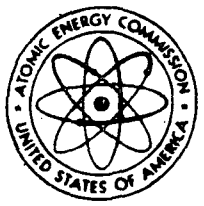
May 1974



U.S. ATOMIC ENERGY COMMISSION

REGULATORY GUIDE

DIRECTORATE OF REGULATORY STANDARDS

REGULATORY GUIDE 5.23

IN SITU ASSAY OF PLUTONIUM RESIDUAL HOLDUP

A. INTRODUCTION

Part 70, "Special Nuclear Material," of Title 10 of the Code of Federal Regulations requires licensees authorized to possess more than one kilogram of plutonium to calculate a material balance based on a measured physical inventory at intervals not to exceed two 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 Regulatory 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. Whenever possible, process equipment should be designed* and operated so as to minimize the amount of holdup. In this guide, procedures are detailed for the in situ assay of the residual plutonium holdup.

Assay information can be used in one of two ways:

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

2. When the limit of error of Pu holdup is not compatible with constraints on the overall LEMUF, the information obtained in the holdup survey can be used to locate principal Pu accumulations and to assure 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 LEMUF 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 glove box walls and floors, accumulate deposits of plutonium which 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 amount of plutonium holdup may be significant in the context of the tolerable facility MUF.

The measurement procedures detailed in this guide are based on the controlled observation of gamma rays and neutrons which are spontaneously emitted by the plutonium isotopes. Because the gamma rays of interest are emitted by Pu-239, gamma ray assay is the preferred

*Design features to minimize holdup in process equipment are the subject of a series of regulatory guides.

assay method whenever its acceptance criteria are satisfied. To accomplish either gamma ray or neutron assay, it is essential to consider the facility in terms of a series of zones which can be independently assayed. Such zones are designated as "collection zones."

1. Delineation of Collection Zones

Typical plutonium process facilities comprise a number of interconnected glove boxes which 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 glove box lines. Finally, storage areas for feed, scrap and waste, and final product are also often located in close proximity to the plutonium process area.

Each facility can be divided into a series of collection zones on the basis of a logical understanding of process activities. Individual glove boxes can be subzoned to improve assay performance, but for most applications, individual glove boxes are examples of suitable size areas for discrete collection zones.

Gamma ray assay for plutonium holdup measurement is practical when a collection zone consists of a single structure of relatively uniform cross section. When a collection zone contains an item of equipment having significant shielding properties and capable of contributing to the holdup, the uncertainty in the holdup prediction based on the observed response may become primarily due to attenuating the radiations in the internal structure. In such cases, neutron assay is applicable.

2. Applicable Methods and Instruments

Two considerations are critical to the selection of methods and instruments. First, to perform an assay, the plutonium radiations 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 developed around penetrating radiations and the detector is collimated to provide for sufficient directionality in the response to resolve a collection zone from its neighbor zones and from the background.

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[1,2] 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 one or two 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 emission rate of gamma rays, a detection sensitivity of less than one gram is generally attainable.

The most useful portion of the spectrum for holdup assay is the Pu-239 gamma ray complex in the 375-440 keV range. The yields of these lines are given in Table B.1.

Table B.1

PROMINENT GAMMA RAYS FROM Pu-239 in ENERGY RANGE 375-440 keV

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

2.1.1 Gamma Ray Detection Instruments

Gamma ray detection systems consist of a scintillation or semiconductor detector sensitive to gamma rays and appropriate electronics.[3] Required electronics include at least a single-channel analyzer and a timer-scaler unit. A second single-channel analyzer used to determine the background radiation correction is a time-saving feature. Battery powered systems are commercially available and can provide operational convenience, particularly in this application.

The detection efficiency and resolution of good NaI(Tl) detectors is generally adequate for this application. CdTe, Ge(Li), and intrinsic Ge detectors have better resolution than NaI(Tl) but cost more, are generally less available, and are more difficult to operate.

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 375-440 keV complex. If the U-237 is in equilibrium with Pu-241, the intensity of this gamma ray is 1.15×10^6 γ/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 440 keV.

Detector dimensions are selected to provide a high probability for detecting the appropriate gamma rays. The geometric detection efficiency increases as the square of the detector radius; however, the weight of the gamma ray shielding material required to collimate the detector also increases when larger detectors are used. The crystal depth is chosen such that most of the gamma rays of interest will lose all their energy within the crystal.

To reduce the pile-up of low energy radiations, the crystal face can be covered with an appropriate shield (e.g., 0.075 cm cadmium). This procedure will reduce counter dead time effects without significantly affecting assay results.

2.1.2 Collimators for Gamma Rays

A shaped shield constructed of any dense material is appropriate for gamma ray collimation. For cost, availability, and ease of fabrication, lead is recommended. Less than 2% of all 400 keV gamma rays striking a 1.5-cm-thick sheet of lead will pass through without having suffered 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.[4] Making this distance variable to reproducible settings will permit adjustment over a range of collection zone sizes.

2.1.3 Check Source for Gamma Ray Assay

It is important to check the operation of the detection system prior to 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 performance 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 Pu) 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.

2.1.4 Calibration Source for Gamma Ray Assay

To calibrate a collection zone, the observed assay response is compared to the response obtained when the zone contains a known amount of plutonium.

Because of the complexity of the assay, the response is assumed to be linear. To be representative of typical holdup situations, the calibration standard is prepared as an encapsulated disk with a bed thickness of less than 0.2 cm. Care must be exercised in the preparation of the calibration standard to ensure that the amount encapsulated of total plutonium, Pu-239, and the amount of Am-241, is known. It is important to measure the gamma ray attenuation through the encapsulating material and correct the calibration standard response to compensate for that attenuation. The amount of plutonium encapsulated in the gamma ray calibration standard is selected to be representative of typical accumulations.

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. Glove box 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, once 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, 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 standard material neutron production rate. 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 and to collimate the detector so that the only neutrons being counted are emanating from the collection zone under assay.

Holdup assay is performed under in-plant conditions where ruggedness, high detection efficiency, and high (γ,n) rejection performance in the detectors is 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 can be connected in parallel to feed a common preamplifier.

He-3 and BF_3 detectors have efficiencies which increase as the energy of the neutrons decrease. To take advantage of this characteristic, the detectors can be surrounded by a neutron moderating material (see Figure B-1). Polyethylene is recommended. The thickness of the moderator is important. When the moderating distance is short, a fraction of the higher energy neutrons pass 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 one 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 1¼ inch when one-inch-diameter tubes are used, and approximately one inch when two-inch-diameter tubes are used.

To shield the detector from low-energy neutrons which may produce a complicated response pattern, the moderator material is covered with a thermal neutron absorber. Cadmium sheeting approximately 0.075 cm thick can 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 Cd cover. For each six 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 B-1.

The weight of the combined detector and collimator assembly can easily exceed requirements for a hand-held detector probe.[4] For this reason, and to provide for reproducible positioning at each assay, a sturdy cart housing both the detector/collimator and the associated electronics is recommended. Further, as the items to be assayed will be at different heights, the ability to raise and lower the assembly to reproducible settings is recommended to expedite the assay and reduce the possibility of errors.

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 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 Pu) into the face of a plug of neutron moderating material (see Figure B-2). The plug is fabricated to fit and close the collimator channel.

2.2.4 Calibration Source for Neutron Assay

To calibrate a neutron assay collection zone, the observed response is compared to the response obtained when the zone contains an additional known amount of plutonium. Neutron assay is less sensitive to attenuation than is gamma ray assay. It is important to know how much plutonium is encapsulated in the neutron assay calibration standard, and the isotopic composition of that plutonium.

The spontaneous neutron production rate from typical reactor plutonium is significantly less than the production rate of 375-440 keV gamma rays. To provide an adequate response for calibration, it is therefore necessary to encapsulate a larger amount of plutonium in the neutron assay calibration standard.

COLLIMATED NEUTRON DETECTOR ASSEMBLY FOR PLUTONIUM HOLDUP ASSAY

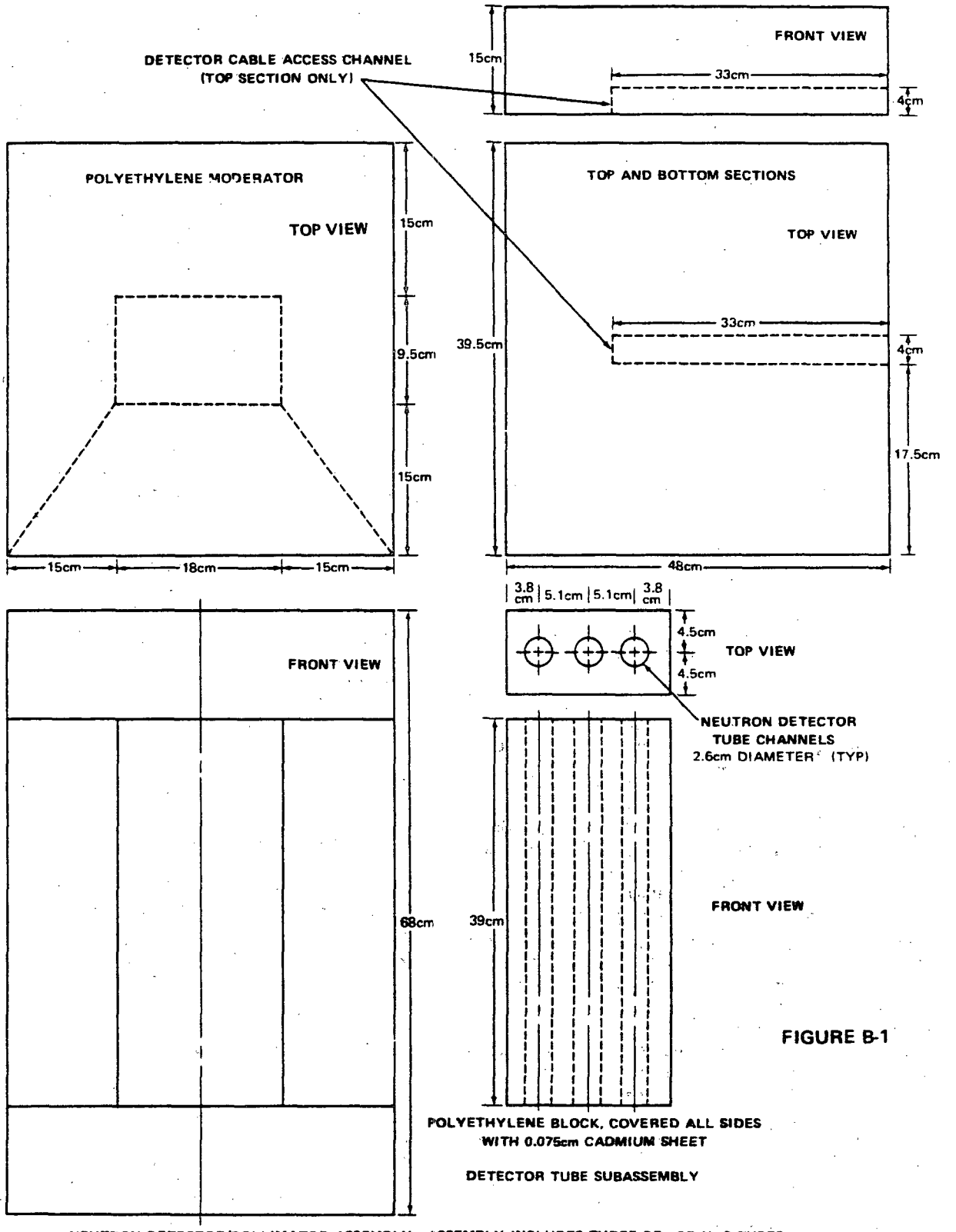


FIGURE B-1

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.

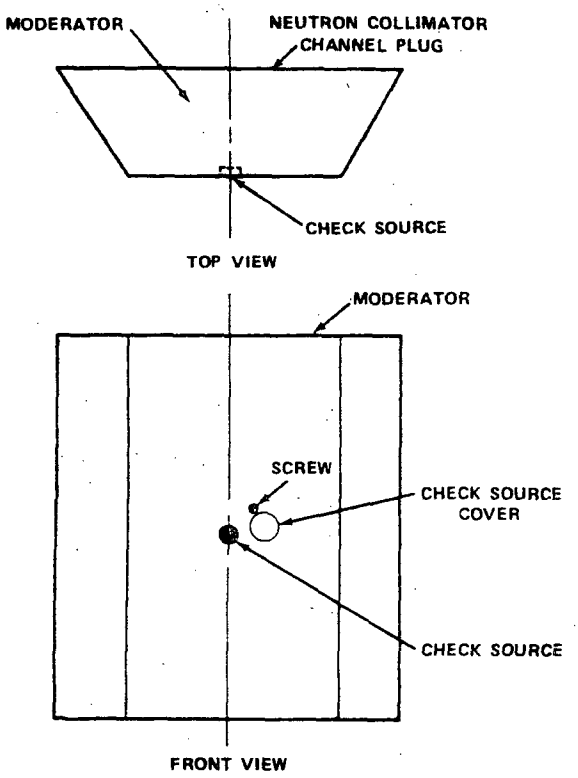


FIGURE B-2 NEUTRON COLLIMATOR CHANNEL PLUG AND CHECK SOURCE

While the amount needed is best determined through an evaluation of typical accumulations, 100 g Pu is adequate for most applications.

The neutron assay 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 of the spontaneous 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. The potentially significant calibration error arising by having too large a neutron yield per gram of plutonium will be corrected in the long term through assay verification tests. In the initial phase of assaying holdup, a rough correction for this yield can be measured by preparing two additional PuO_2 sources containing 1/3 and 2/3 of the neutron assay calibration standard mass. These samples need not be encapsulated, as they will be measured only once and can then be returned to the process stream.

The PuO_2 used in this test is taken from the same batch used to prepare the neutron assay calibration standard. After weighing out the proper quantities, the PuO_2 is put into containers having close to the same geometry as found in the neutron assay calibration standard. Each test sample is transferred to an empty glove box and positioned next to the window for measurement. The neutron assay probe is positioned as close as possible to the sample but outside the glove box. After the measurement is made, that sample is transferred from the glove box and the next sample is transferred in and positioned in the identical location for measurement. A plot of counts minus background as a function of PuO_2 mass is made and the points visually fitted using a French curve. If there is no multiplication, a straight line can be drawn through the origin connecting all points. Multiplication is indicated when the curve turns upward, indicating an increase in counts per gram as the mass of PuO_2 increases. A correction term is obtained by determining the increase in counts per gram at the mass value corresponding to the neutron assay calibration standard mass. This increase is readily determined by plotting the straight line through the origin and the lowest mass sample response and reading the difference in counts between the two lines at the abscissa coordinate corresponding to the neutron assay calibration standard mass. All measurements relating to this standard are thereafter reduced by the ratio of the difference in counts to the observed counts.

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 Sections B.2.1.2 and B.2.2.2. Two additional means exist to further 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 positioning the detector above or below the collection zone, 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 containing ~5 cm of neutron moderator (e.g., benelex, WEP, or polyethylene) and ~0.5 cm lead sheet is recommended, mounted on wheels as an upright panel. To use such a panel, two measurements are required.*

$$R_1 = R_{cz} + R_{Interference} \quad (1)$$

$$R_2 = R_{cz} + TR_{Interference} \quad (2)$$

where

R_1 is the assay response obtained before the shadow shield is moved into position,

R_2 is the assay response obtained with the shadow shield in position,

R_{cz} is the response component attributable to the collection zone under assay,

$R_{Interference}$ is the response component attributable to the interfering radiations, and

T is the transmission through the shadow shield. Note that T represents a measured transmission—T_γ for gamma rays or T_n for neutrons. T_γ and T_n are measured by counting radiations from any arbitrary source of plutonium with the shield between the source and detector and again with the shadow shield removed:

$$T_\gamma = (R_\gamma) \text{ shield in}/(R_\gamma) \text{ shield out} \quad (3)$$

$$T_n = (R_n) \text{ shield in}/(R_n) \text{ shield out} \quad (4)$$

To correct for the interference, subtract R_2 from R_1, and solve for $R_{Interference}$:

$$R_{Interference} = \frac{(R_2 - R_1)}{(1 - T)} \quad (5)$$

To ensure that this correction is sufficiently accurate, it may be necessary to extend the length of the normal counting period to accumulate sufficient counting statistics (1% statistics are generally adequate for this application).

4. Calibration of Collection Zones

Each collection zone is independently calibrated, as background factors and the composition of each zone vary widely from zone to zone. A collection zone is best calibrated through the in situ measurement of known calibration standards. When such a program is not possible, the calibration can be based on the calculation of the anticipated response or through measuring a mockup of the collection zone of interest.[5]

*Response terms refer to neutron or gamma response, as appropriate.

The calibration obtained through this procedure is recommended until a history of comparisons between predicted and recovered holdup quantities is developed, as described in Section B.5 of this guide.

4.1 Detector Positioning

To calibrate each collection zone, the best position or series of positions is selected to observe the collection zone with the least amount of interference from principal structural components. It is important to view the collection zone with the detector located between the collection zone and all areas used for Pu storage during inventory. A three-dimensional approach can be investigated, positioning the detector on top of or below the collection zone if it is not possible to have an unobstructed, interference-free side view of the collection zone. The use of shadow shielding can be explored if it is not possible to get a clear view of each collection zone for assay.

On the basis of a detailed examination of the physical layout of the facility, some preliminary measurements are made to determine optimum detector positions for holdup assay. Once the assay positions for the detector and shadow shields are established, permanently marking the assay positions will facilitate subsequent measurements.

4.2 Calibration Sources

Since this assay is to measure the amount of plutonium holdup, it is appropriate to use plutonium as the calibration standard material. Further, as the plutonium holdup will generally be distributed over a large surface area, it is recommended that the gamma ray calibration standard be fabricated to resemble this characteristic, as described in Section B.2 of this guide.

4.3 Calibration Procedures

Once the principal items containing plutonium have been removed and the detector located in its assay position, the response from a calibration standard combined with the plutonium already held up is obtained. When the collection zone is appropriately isolated, two factors influence the observed response from the calibration standard:

1. the location of the calibration standard within the collection zone, and
2. the shielding of radiations from the calibration standard caused by the items comprising the collection zone.

The geometric response variation is measured by observing the response from one calibration standard with the other standard removed from the collection zone under investigation. The calibration standard response is measured with the standard positioned in various parts of the collection zone, avoiding internal items which may attenuate the radiation emanating from the standard.

When neutron assay is employed or when the collection zone consists of a hollow box, pipe, or duct, attenuation is either relatively uniform or negligibly small. The calibration of each collection zone then becomes a matter of appropriately averaging the geometric response variations. The average response of the entire collection zone is assumed to properly represent that zone. If, however, it is known that plutonium accumulates in one particular location within a collection zone, the response of the standard is emphasized when located near the principal collection site.

If the item to be assayed consists of a large unit, assay performance may be enhanced by subdividing the unit into smaller contiguous measurement zones. The repeat dimensions of the subzones are determined by measuring the response while moving the standard along an axis perpendicular to the detector centerline. By studying the response curve, the distance D is selected as the point beyond which sufficient activity is detected to flatten the response within the subzone. Each subzone will measure 2D across its face. An example is illustrated in Figure B-3. As the response about the centerline is assumed to be symmetrical, only half of the traverse is indicated. In Figure B-3, D is selected such that the area under the curve to the right of D is approximately equal to the area above the curve to the left of D (Area $A_1 \simeq$ Area A_2). *Note:* the distance from the collection zone to the detector or the distance from the crystal face to the end of the collimator, or both, can be varied to divide the collection zone into an integral number of subzones.

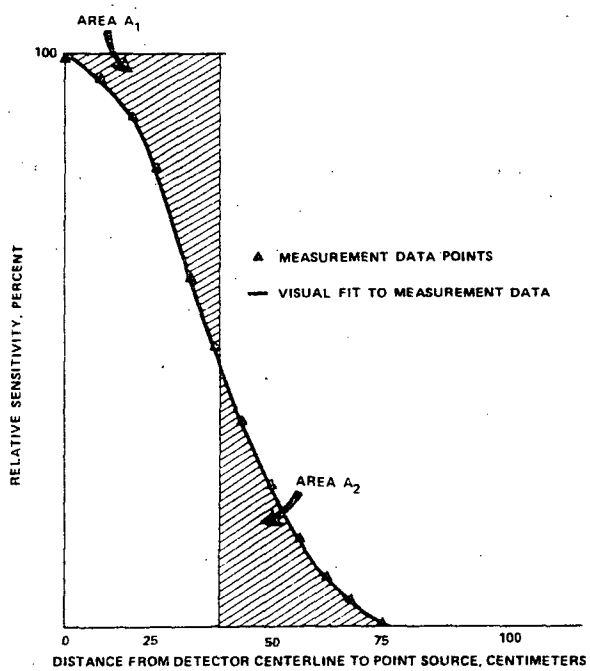


FIGURE B-3 EQUIVALENT DIAMETER SUBZONE TO ACHIEVE A FLAT PLANAR RESPONSE. SELECT D SUCH THAT AREA A_1 = A_2.

To use this relationship, the detector is first positioned at point d and a reading is taken. Point d is the center of the first subzone, selected to coincide with the physical edge of the calibration zone. The detector is then moved a distance 2D along the traverse to the center point of the second subzone, and the second measurement taken. The cycle is repeated to include all of the larger collection zone. The value interpreted for calibration for each subzone corresponds to the maximum of the traverse across each subzone because the response has been flattened. The content of the entire collection zone is the sum of the contributions from the subzones.

5. Estimation of the Holdup Error

The overall uncertainty associated with the measured plutonium holdup is due to (1) the uncertainty in the observed response and (2) the uncertainty in the interpretation of that response. The random uncertainty components in this application are frequently negligible in comparison with the geometric uncertainty and the uncertainty in the isotopic composition. In this assay application, it is appropriate to estimate the assay error components by assuming the measured range (R_i) of the ith fluctuation constitutes an interval four standard deviations wide. The midpoint of the range estimates the mean effect, and the distance from the midpoint to each extreme comprises an estimated 95% confidence interval. The error attributable to this effect is then approximately

$$\sigma_i^2 = \left(\frac{R_i}{4}\right)^2 \qquad (6)$$

If a severe effect is noted, the response can often be corrected for the variation in the corresponding parameter by measuring the value of that particular parameter at the time of the assay. Using a measured relationship between the response and the value of that parameter, the observed response is corrected.

5.1 Response Uncertainties

5.1.1 Counting Statistics

The magnitude of the uncertainties attributable to variations in the geometric distribution and in the attenuation of the radiations are expected to dominate the total response uncertainty. The relative standard deviation due to counting statistics can usually be made as small as desired through (1) using more efficient detectors or (2) extending the counting period. Having 1000 to 10,000 net counts is generally sufficient for most holdup assay applications.

5.1.2 Instrument Instabilities

Fluctuations in ambient temperature, humidity, electronic noise, and line voltage (for

non-battery-powered electronic units) generally affect the stability of electronic systems. The magnitude of this uncertainty can be estimated by monitoring the check standard response and determining the range of variability as described in Section B.5 of this guide.

5.1.3 Geometric Uncertainty

The geometrical variation in the observed response is measured by moving the calibration source within the bounds of each collection zone. Two cases are described below.

5.1.3.1 Isolated Collection Zones

When a single unit comprises a collection zone, the standard is moved to all sites within the zone at which an accumulation of plutonium might occur. With sufficient collimation, the response for the collection zone under investigation is independent of its neighbor zones. The average of the response, weighted to reflect prejudgments on the likelihood of accumulation sites, is then used as the calibration point. As shown in Section B.5, the range of values can be assumed to comprise an expectation interval four standard deviations wide. The geometric error is then estimated using Equation 6.

5.1.3.2 Overlapping Collection Zones

When a collection zone is subdivided into overlapping subzones, the geometric uncertainty due to the dimension perpendicular to the detector collection zone centerline is eliminated through the area-averaging calibration method described in Section 4.3.

The uncertainty in the depth dimension in each subzone can be determined through the procedure outlined for isolated collection zones. Judgment can be used to weight the calibration data to emphasize principal accumulation sites.

5.1.4 Attenuation Uncertainty

If the attenuation is not extreme, it can be measured in situ, mocked up, or computed for the different conditions encountered. The worst and best cases can be assumed to determine the range of permissible effects. Using Equation 6, the magnitude of this uncertainty component can then be estimated. Again, judgment is appropriate to weight the correction factor.

5.2 Interpretation Uncertainties

Two factors are central to the issue here, assuming that the calibration standard material is similar to the held-up material.

5.2.1 Interfering Radiations

5.2.1.1 Gamma Ray Assay

An uncertainty in the observed gamma ray response may arise due to the presence of extraneous gamma ray emitters or due to fluctuations in the background from the Compton scattering of higher-energy gamma rays. The shape of the background gamma ray spectrum may change in such cases to such an extent that even with the energy windows stabilized, the background correction is irregular and uncertain. The magnitude of this effect is generally small. It can be monitored by observing the spectrum with a multichannel analyzer, but unless the data on periodically recovered holdup accumulations are in error, this contribution can be ignored.

5.2.1.2 Neutron Assay

A change in the neutron yield for a plutonium sample of fixed isotopic content is primarily attributable to the fluctuation in the concentration of high (α,n) yield impurities.* Judgment can be used to determine the range of permissible impurity concentrations. The variation in a typical neutron yield can then be predicted using the methods discussed in the Appendix of this guide. Again, the range of permissible variations is assumed to constitute an acceptance interval from which the component error is computed using Equation 6.

5.2.2 Isotopic Uncertainties

If the process equipment is cleaned each time the isotopic composition of the plutonium feed is varied, the holdup will consist primarily of the current material. New calibration standards can be prepared or the previous yield data can be normalized using the methods presented in the Appendix to correct for this effect. When mixing occurs, use of the stream-averaged isotopic composition is appropriate. The uncertainty bounds are estimated by considering the highest and lowest fissile isotopic batches and computing the corresponding range.

5.3 Holdup and Its Associated Error

The amount of Pu holdup can be measured through the systematic application of the program developed in conjunction with the principles and pitfalls discussed herein. For each collection zone, measured holdup and its error can be determined.

*Over a long period of time the α-particle production rate increases due to the ingrowth of Am-241.

5.3.1 Initial Operations

During the initial phase of operations, the error associated with the in situ assay of plutonium holdup is estimated by combining the component errors determined in the preceding sections of this guide (B.5.1 and B.5.2).

5.3.2 Routine Operations

To ensure the validity of assay predictions and to more realistically estimate the uncertainty in those predictions, it is necessary to establish a program to measure the amount of plutonium recovered when a collection zone is cleaned out. By comparing the amount of plutonium recovered to the recovery amount predicted, the collection zone calibration can be updated and the assay error can be based on relevant verification tests.

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

$$(Pu)_{assay} = R_{before} - R_{after} \quad (7)$$

The difference (Δ) in assay and recovery,

$$\Delta = (Pu)_{assay} - (Pu)_{recovery} \quad (8)$$

is then computed.

The standard deviation in the Δ values (s_Δ) is computed separately for each collection zone, including no more than the twelve preceding measurement tests:

$$s_\Delta = \left[\left(\sum_{i=1}^{K} \Delta_i^2 - \left(\sum_{i=1}^{K} \Delta_i\right)^2 /K\right) / (K-1)\right]^{1/2} \quad (9)$$

When a value of Δ is determined, it is used to update the estimate s_Δ. The standard deviation estimate s_Δ can be used to estimate the error in the assay prediction for the collection zone for which it has been established.

The amount of plutonium collected during the cleanout of a specific collection zone can be assayed through sampling and chemical analysis, through calorimetry, or through other applicable nondestructive assay methods (e.g., spontaneous fission coincidence detection or gamma ray assay). Each of these topics is the subject of a Regulatory Guide.

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.

Note: Care must be exercised during the fabrication and use of check sources and calibration standards to ensure their continued integrity and to prevent contamination.

1. Delineation of Assay Collection Zones

A plan of each plutonium processing facility should be examined to establish independent collection zones. Individual glove boxes and similar containment structures should be so identified. Using the layout and touring the facility, an assay site(s) for each collection zone should be selected:

1. Assay site(s) should afford a clear, unobstructed view of the collection zone with no other collection or storage areas in the line of sight of the collimator assembly. Location of the detector probe above or below the collection zone should be considered if an unobstructed side view is not possible. If an unobstructed view is not possible, shadow shielding should be used to isolate the collection zone for assay.

2. The assay site should be set back as far as possible from each collection zone to reach a compromise between interference from neighbor zones and efficient counting.

3. Gamma ray assay should be applied to measure the plutonium held up in all collection zones containing less than the neutron detection limit and for single containment structures which do not contain irregularly shaped structural components capable of significantly attenuating the emerging gamma rays. Neutron assay should be applied to measure the accumulation of plutonium holdup in all structures not suitable for gamma ray assay.

4. Each collection zone should be uniquely numbered. (Neutron collection zones could be preceded by an "N", gamma ray collection zones by a "G". Subzones should be identified by an alphabetic suffix to the collection zone identification.)

5. Each assay site should be marked with paint or colored tape on the floor. (To be consistent, blue tape should be used for neutron assay sites, orange for gamma ray sites.) The height setting for midpoint assay should be recorded in the measurement log corresponding to each assay site.

2. Assay Instruments

Neutron and gamma ray assay capability should be provided using separate or compatible electronics with interchangeable detector probes. Compatible electronics

should 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% per °C. Battery-powered electronics should be provided to 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 440 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 should have FWHM resolution equal to or better than 7.5% at 662 keV (Cs-137 gamma ray). NaI(Tl) can meet such specifications and is 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 one inch. A two-inch depth is recommended.

The crystal should be stabilized with a suitable radioactive source. An internal CsI 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 to 0.150 cm cadmium sheet 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 390 keV to 440 keV unless equilibrium of the U-237 and Pu-241 can be assured. The 333-keV region of the gamma ray spectrum should be excluded. With NaI detectors, it is necessary to exclude the 375 keV gamma ray to ensure that the tail from the 333 keV complex is not added. The second channel should be set above the first window to provide a background correction for the assay window. This second window should be set from approximately 450 keV to 600 keV.

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 extendible over the end of the crystal to reproducible settings to vary the degree of collimation for different collection zones.

2.1.3 Gamma Ray Check Source

To ensure the continued normal operation of each system, an encapsulated plutonium check source should be provided. The source should be small enough to be implanted in a section of shielding material so shaped as to close off the collimator opening. The check source should be positioned adjacent to the detector. The source should contain an amount of plutonium sufficient to provide a gross count rate of 1000 to 10,000 counts per second.

2.1.4 Gamma Ray Calibration Source

To permit the calibration of gamma ray assay collection zones, a calibration standard should be fabricated by encapsulating plutonium oxide in a disk. The isotopic composition of the plutonium and the abundance of Am-241 should be measured and be chosen to be nominally representative of the plutonium being processed. The total amount of plutonium encapsulated should be closely monitored. Attenuation losses within the bed of PuO_2 and through the encapsulating material should be measured and the calibration standard response normalized to counts per gram incorporating these corrections.

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 obtainable from a single detector tube. 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. A recommended configuration is diagrammed in Figure B-1.

2.2.2 Neutron Collimator

A slab collimator or concentric cylinder collimator of a suitable neutron moderator material (e.g., polyethylene) should be constructed to completely surround the detector with its associated moderator and filter assembly, leaving open only the collimator channel. A recommended configuration is shown in Figure B-1.

The moderator thickness should be selected to provide the directionality required for each facility. A directionality profile providing a 10:1

response ratio (six inches of polyethylene) should be adequate for most applications; however, each situation should be evaluated as discussed in Part B of this guide.

2.2.3 Neutron Check Source

Any neutron source which emits approximately 100-10,000 neutrons/second is acceptable for this application. The source should be small enough to be contained within a section of neutron moderator material so shaped as to completely fill the collimator channel of the detector assembly. The source should be implanted directly adjacent to the neutron detectors, outside the cadmium thermal neutron filter. A recommended configuration for this assembly is diagrammed in Figure B.2.

2.2.4 Neutron Assay Calibration Standard

To permit the calibration of neutron assay collection zones, a calibration standard should be fabricated by encapsulating PuO_2. The PuO_2 should be nominally representative of the plutonium being processed in isotopic composition, in Am-241 content, and in the content of high (α,n) yield target materials. The amount of plutonium to be encapsulated should be chosen to be representative of the amounts of plutonium estimated to be held up in typical neutron assay collection zones.

The neutron yield of the calibration standard should be measured and also computed using the method described in the Appendix. The observed neutron count rate should be normalized.[6] If the predicted response differs by more than 10%, the response should be normalized as discussed in Section B.2.2.4.

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.

2.4 Notation of Operating Parameters

When compatible electronics are used to facilitate neutron and gamma ray assay, a notation of the respective settings should be affixed to the electronics unit. To decrease the likelihood of incorrect settings, the neutron probe and the appropriate electronics settings should be color-coded blue; the gamma ray probe and corresponding electronics settings should be coded orange.

3. Calibration

Each collection zone should be independently calibrated when all in-process material has been located so that the response from the calibration standards will not be influenced by the in-process material.

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.

3.2 Zone Calibration

The geometric response profile for each collection zone should be determined by measuring the variation in the response as a calibration standard is moved within the defined limits of the collection zone. The response variation should then be averaged to determine the response per gram of plutonium for that collection zone. The averaging should be weighted to reflect known local accumulation sites within each collection zone. The response per gram should be used to directly translate the observed response to grams of plutonium, after the response is corrected for background.

3.2.1 Subzone Calibration

When a collection zone is too large to be accurately measured in a single assay, the collection zone should be divided into overlapping subzones. The repeat dimensions of each subzone perpendicular to the detector-to-collection-zone line should be determined so that the response variation across that distance is nulled. Using this procedure, the residual geometric uncertainty should be determined by measuring the response as a calibration standard is moved along the depth coordinate. The calibrated response should then reflect the average of the depth response, weighted to reflect known accumulation sites.

4. Assay Procedures

4.1 Assay Log

An assay log should be maintained. Each collection zone or subzone should have a separate page 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 plutonium from the calibration in addition to position and instrument electronic setting verification.

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.

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 from the process areas to approved storage areas to minimize background radiations.

4.3 Measurements

The assay cart should be moved in sequence to the assay site(s) corresponding to each collection zone. Assaying all gamma ray sites before assaying neutron sites (or vice versa) is recommended.

Before assaying each collection zone, the operator should verify the floor location, probe selection, probe height, and electronics settings. All check and calibration sources should be sufficiently removed 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 assure 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 assure compliance for each collection zone.

Having met all preceding requirements, the measurement at each site should be taken, recorded, and converted to grams plutonium. If each value is within an expected or permissible range, the cart should be moved to the next site and the cycle repeated. If a high response is noted, the cause should be investigated. If the collection zone contains an unexpectedly large content of plutonium, 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 recovered material quantity used to test the validity of the zone calibration.

5. Estimation of the Holdup Error

During the initial implementation of this program, the error quoted for the holdup assay should be computed on the basis of estimating the error components, as described in Sections B.5.1 and B.5.2.

Prior to the cleanout of any collection zone for whatever purpose, that zone should be prepared for assay and measured as described in Section C.4 of this guide. Following this assay, the collection zone should be cleaned out and the collected plutonium should then be assayed using an appropriately accurate assay method. When the collection zone has been cleaned and the collected plutonium removed, the collection zone should be reassayed. The recovered plutonium should be used to update the calibration and, from the sixth test on, should serve as the assay error estimate. Separate records should be maintained for each collection zone to estimate the error in assaying the plutonium holdup.

To ensure that error predictions remain current, only data of the twelve preceding independent tests should be used to estimate the assay error. Collection zones not cleaned for other purposes should be cleaned for assay verification at intervals not to exceed two months.

REFERENCES

1. R. Gunnink and R. J. Morrow, "Gamma Ray Energies and Absolute Branching Intensities for $^{238,239,240,241}Pu$ and ^{241}Am," UCRL-51087 (July 1971).
2. J. E. Cline, R. J. Gehrke, and L. D. McIsaac, "Gamma Rays Emitted by the Fissionable Nuclides and Associated Isotopes," ANCR-1069 (July 1972).
3. L. A. Kull, "Catalogue of Nuclear Material Safeguards Instruments," BNL-17165 (August 1972).
4. An example of a collimator for uranium gamma ray assay is found in R. B. Walton, et al, "Measurements of UF_6 Cylinders with Portable Instruments," Nucl. Technol., *21*, 133 (1974).
5. W. D. Reed, Jr., J. P. Andrews, and H. C. Keller, "A Method for Surveying for Uranium-235 with Limit of Error Analysis," Gulf-GA-A12641 (June 1973).

APPENDIX

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 created through induced fission of Pu-239 or Pu-241. The total neutron yield per gram of plutonium holdup will then be the sum of the spontaneous fission and (α,n) contributions:

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

1. Spontaneous Fission Neutrons

To determine the spontaneous neutron yield per gram of plutonium held up within a collection zone, the isotopic composition of the plutonium and uranium (if present) must be known. The contribution from spontaneous fission can generally be calculated by neglecting the contribution from U-238:

$$Y_{SF} = W_{238}Q_{238} + W_{240}Q_{240} + W_{242}Q_{242} \quad (2)$$

where

W_i = weight fraction of the ith plutonium isotope. For reactor fuel applications, $W_{238} + W_{239} + W_{240} + W_{241} + W_{242} \equiv 1$

Q_i = spontaneous fission neutron yield per gram of the ith plutonium isotope (see Table 1).

2. (α,n) Neutrons

The major contribution to the total neutron production from (α,n) reactions will typically be due to the O-18 (α,n) Ne-21 reaction when the plutonium exists as the oxide. The yield from this reaction per gram of plutonium can be calculated using the isotopic weight fractions (W_i) and the Y_i yield data given in Table 1.

$$Y(\alpha,n)\ Oxy = \sum_i W_i Y_i \quad (3)$$

The yield per gram of PuO_2 is calculated by multiplying the yield per gram of plutonium by the gravimetric dilution factor ($Pu/PuO_2 \cong 0.882$).

The presence of certain impurities can contribute substantially to the total (α,n) production rate. Approximate values of (α,n) impurity yields for the highest yield (α,n) target materials are given in Table 2. To compute the impurity (α,n) contribution, the total α particle production is determined. Production rates of α particles per gram of the principal nuclides of interest are shown in Table 1. This contribution to the total neutron yield can be computed using the relationship:

$$Y(\alpha,n)\ Impurity = Y_\alpha \sum_j P_j I_j \quad (4)$$

TABLE 1
α Particle and Spontaneous Fission Neutron Yields

Nuclide	Half-life (yr)	Alpha Activity (α/sec-gram)	$PuO_2(\alpha,n)$ Yield[a] (n/sec-gram)	Spontaneous Fission (n/sec-gram)
Pu-238	87.78	6.33×10^{11}	1.71×10^{4}	2.57×10^{3}
Pu-239	24,150	2.30×10^{9}	54.5	2.22×10^{-2}
Pu-240	6,529	8.43×10^{9}	202.1	1.03×10^{3}
Pu-241	14.35[b]	9.39×10^{7}	2.03	2.43×10^{-2}
Pu-242	379,000	1.44×10^{8}	3.13	1.75×10^{3}
Am-241	433.8	1.27×10^{11}	3.46×10^{3}	6.05×10^{-1}
U-234	2.47×10^{5}	2.29×10^{8}	4.65	5.67×10^{-3}
U-235	7.1×10^{8}	7.93×10^{4}	1.37×10^{-3}	5.96×10^{-4}
U-238	4.51×10^{9}	1.23×10^{4}	1.93×10^{-4}	1.12×10^{-2}

a – Oxygen yield from PuO_2 form only.
b – α-branching ratio = 2.46×10^{-5}

where

Y_α = total α production

$$= \sum_i W_i \alpha_i + W_{Am} \alpha_{Am}$$

W_i = Pu isotopic weight fractions

W_{Am} = Am weight fraction = Am/Pu

α_i = α yield per gram of nuclide i (see Table 1)

TABLE 2
(α,n) Yield Rates of Low-Z Impurities in PuO_2[a]

Impurity	P_j (n/α-ppm)
Li	6.29×10^{-12}
Be	2.00×10^{-10}
B	4.63×10^{-11}
C	2.77×10^{-13}
O[b]	1.56×10^{-13}
F	2.44×10^{-11}
Na	3.00×10^{-12}
g	2.67×10^{-12}
Al	1.45×10^{-12}
Si	3.25×10^{-13}

[a]Assumes zero yield from all other impurities.
[b]Oxygen not contained in oxide.

P_j = (α,n) yield per ppm of the impurity j (see Table 2)

I_j = impurity j content, expressed in ppm (weight) of plutonium.

3. Sample Calculation (PuO_2-UO_2)

Consider the case of recycle plutonium blended to 3 wt % Pu in a normal UO_2 matrix, where the isotopic composition is Pu-238 (.25%), Pu-239 (75.65%), Pu-240 (18.48%), Pu-241 (4.5%), Pu-242 (1.13%), and Am-241 (.28% of Pu).

For mixed oxides, the oxygen density is approximately the same for the case of PuO_2. This fact, together with the atomic similarity of uranium and plutonium, justifies the assumption that the oxygen (α,n) yield per gram of mixed oxide is the yield per gram of PuO_2, further reduced by the blending ratio, Pu/(Pu + U).

Using the values given in Table 1, the spontaneous fission yield and total α production per gram of plutonium can be computed. Results are shown in Table 3.

The α particle yield of plutonium is constant in time for all intents. However, the Am-241 α production increases at a rate which results in approximately a 0.3% increase per month in the total α production, for the range of plutonium isotopic compositions intended for reactor fuel application.

In the present example, the impurity levels of the principal (α,n) target materials are shown in Table 4. The neutron yields attributable to (α,n) interactions on those

TABLE 3
Sample Calculation

Nuclide	W	Spontaneous Fission (n/sec-g Pu)	Alpha Production (α/sec-g Pu)	PuO_2(α,n)[a] (n/sec-g Pu)
Pu-238	.0025	6.4	1.58×19^9	42.6
Pu-239	.7565	<.05	1.74×10^9	41.3
Pu-240	.1848	189.4	1.56×10^9	37.3
Pu-241	.0450	<.05	4.23×10^6	0.1
Pu-242	.0113	19.8	1.63×10^6	<0.05
Am-241	.0028	<.05	3.56×10^8	9.7
Total Yields		215.6	5.26×10^9	131.0

[a] -- oxygen yield only.

impurities are also shown in Table 4, calculated using the α particle production rate of 5.3×10^9 α/sec-g Pu, computed above. In this example, the mixed oxides are composed of blended PuO_2 and UO_2 particles approximately 40 microns in diameter. If the particle size were smaller or the mixed oxide was created through coprecipitation, the uranium impurity content would also contribute to the plutonium (α,n) yield. This contribution can be ignored for large particles and estimated by combining the impurities for small particles and coprecipitated oxides.

The total neutron yield in this example is 380 n/sec-g Pu. In this example, the percentage of plutonium to the total Pu + O is 0.8835. Using this gravimetric dilution factor, the neutron yield is 336 n/sec-g PuO_2. If the PuO_2 is blended with UO_2 to 3%, i.e., $PuO_2/PuO_2 + UO_2 = 0.03$, the neutron yield from the blend will be 10.1 n/sec-g MO.

TABLE 4
Impurity (α,n) Yield

Impurity	Arbitrary Concentration (ppm)	(α,n) Yield (n/sec-g Pu)
Li	9	0.30
Be	8	8.42
B	10	2.44
C	200	.30
F	125	16.0
O[a]	4600	3.77
Na	120	1.90
	Total	33.1

[a]Oxygen present in moisture, not as oxide.