

5. NONDESTRUCTIVE ASSAY OF HOLDUP

T. Douglas Reilly

I. INTRODUCTION

The term "holdup" refers to the nuclear material deposited in the equipment, transfer lines, and ventilation systems of processing facilities. Reprocessing, fuel fabrication, conversion, and enrichment require very large facilities that can contain many pumps, ovens, centrifuges, filters, diffusers, and hundreds of kilometers of pipes and ducts. During years of operation, significant quantities of uranium and/or plutonium can build up in this equipment. Operators need to know the location and amount of holdup for reasons of accountability, criticality safety, radiation safety, waste management, and efficient plant operation. Sometimes the term holdup is also applied to in-process inventory, if this must be known for verification or accountability purposes. Holdup is difficult to measure and while it is usually a small fraction of plant throughput, it can often amount to many kilograms of nuclear material and this limits the accuracy of the nuclear material balance within the facility. A diverter could, in principle, remove one or more significant quantities (SQ) of HEU or plutonium and hide the diversion as unmeasured holdup deposits within the plant. IAEA safeguards inspectors rarely attempt to measure holdup; although they have participated with Los Alamos in a holdup measurement campaign at the Ulba Fuel Fabrication Plant in Ust-Kamenogorsk, Kazakhstan. Reference 1 presents an excellent discussion of holdup measurements.¹

Holdup measurements must cover a range of material types as shown in Table 1 that lists some of the items measured during a holdup campaign at an HEU processing facility. Process history determines which materials may be deposited. The range of deposit thickness, presence of different material types (isotopic mixtures), and chemistry influence holdup measurements. The range of ²³⁵U enrichment in some facilities includes depleted (0.3%) up to 97%, and that of ²⁴⁰Pu at other facilities ranges from 2% to 45%. Such a range of materials is usually not found in commercial nuclear facilities. Because the equipment in large facilities is extensive, the total holdup may be large, even if deposit thicknesses are small.

Holdup measurements are usually made using gamma-ray techniques, although neutron measurements are also used. There is some experience using thermoluminescent dosimeters (TLD) to measure holdup deposits in gloveboxes or heavy equipment where it is difficult to insert gamma-ray detectors. Such dosimeters usually receive most of their dose from x-rays or low-energy gamma-rays, so the results are more susceptible to attenuation or geometry effects than those obtained with gamma-ray detectors. However, measurement performance can be comparable if the TLDs are carefully calibrated using mockups of the equipment to be measured.² Gamma rays have several advantages over neutrons in measuring holdup, because they are easily collimated allowing the locations and distributions of deposits to be defined. The gamma-ray peaks confirm the identities of the nuclides present. Multiple nuclides and elements can be measured independently and simultaneously by choosing the detector and peaks appropriately. Shielded gamma-ray detectors and the required electronics can be small and lightweight so that measurements can be performed in locations that are difficult to access.

Routine NDA measurements are usually conducted on well contained nuclear material items. This is not often the case for holdup measurements, especially those in plutonium processing facilities. Figure 1 shows four views of holdup measurements during the decommissioning of a

plutonium processing facility. The photographs illustrate well some of the difficulties that may arise when measuring holdup.

II. GAMMA-RAY SIGNATURES AND EQUIPMENT

Faced with a mix of material types for plutonium or uranium, the resolution provided by germanium or Peltier-cooled CdTe should be considered if there are possible biases from spectral interferences. When process knowledge is unable to specify isotopics, these high-resolution detectors may be required for preliminary surveys. When isotopic composition is sufficiently well known and interferences unlikely, even low-resolution scintillators such as sodium iodide (NaI) and bismuth germanate (BGO) can make useful holdup measurements. Table 2 lists the gamma-ray peaks commonly chosen to measure the listed nuclides.

Table 1. Example of Items Containing Holdup at Uranium Facility³

Process Component	HEU Content (g)
HEPA filter	< 0.2
Crop Shear	$0.3 < {}^{235}\text{U} < 1.2$
Motor M28619	$15 < {}^{235}\text{U} < 61$
Gear box	$6 < {}^{235}\text{U} < 25$
Furnace B	$13 < {}^{235}\text{U} < 51$
Spare Furnace	$17 < {}^{235}\text{U} < 67$
Lathe	129
Freon Cart	77
Sawbench 1	0.164
cooling hut HEPA filter	17
Exhaust elbow	3.50
outgassing oven	0.105
Cooling hut	17
HEPA filter hut	6.84
Gate Valve	0.85
Pressure cookers	10.8
Exhaust Component	1.42
Riser Crusher	18



Fig. 1. Holdup measurements during Pu facility decommissioning⁴

Table 2. Common Gamma Rays for Holdup Analysis

Isotope	E_γ (keV)	Intensity ($\gamma/\text{g-sec}$)
^{238}Pu	153	5.9×10^6
^{235}U	186	4.32×10^4
$^{241}\text{Pu} - ^{237}\text{U}$	208	2.04×10^7
^{239}Pu	414	3.42×10^4
^{241}Am	662	4.61×10^5
^{238}U	1001	73

If scintillators like NaI or BGO are used, it should be noted that they exhibit a strong gain dependence on temperature. The effective gain of NaI may drop by one to three percent per ten-degree centigrade increase in temperature. A simple and practical stabilization technique is to regularly measure a gamma-ray source to compensate for drift. The 60-keV gamma ray from ^{241}Am ($t_{1/2} = 432.2$ y) is commonly used as a reference peak.

Figure 2 shows the gamma-ray spectrum from low-burnup (93% ^{239}Pu) plutonium measured with four different detectors (NaI, coplanar-grid cadmium-zinc-telluride (CPG CZT), Ge, and Peltier-cooled CdTe). The detector most commonly used for holdup measurements is NaI. A NaI thickness of 1.25 cm absorbs 80% of ^{235}U gamma rays at 186 keV. A thickness of 5 cm absorbs 85% of ^{239}Pu gamma rays at 414 keV. The intermediate-resolution CZT is equal in sensitivity to the 2.5-cm-diameter NaI in spite of its limited size. Cubic CZT crystals as large as 1.5 cm on a side absorb up to 95% and 40% of gamma rays at 186 and 414 keV).

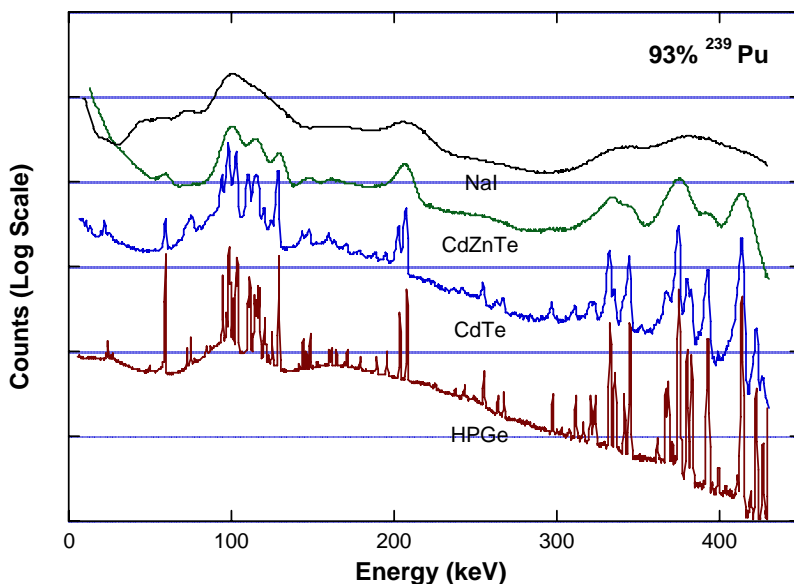


Fig. 2. Plutonium spectrum measured by NaI, CdZnTe, cooled CdTe, and high purity Ge detectors.

Interferences can add unwanted counts to the assay peak. Detectors with improved resolution and peak shape reduce bias from interference. The use of Ge detectors is generally difficult because of their weight. Recent progress with CPG CZT detectors is favorable for portable gamma-ray measurements.⁵ The new solid-state detectors are also more compact, reliable, and stable than scintillators. A large CPG CZT detector can resolve interfering gamma rays from the ^{232}U decay chain that appear in recycled uranium. A gamma ray at 238 keV is produced at the end of this decay chain. It is not resolved from the 186-keV gamma in NaI, but it does not

interfere in Ge or CZT. Gamma-ray peaks from ^{241}Pu - ^{237}U (332 keV), ^{241}Am (323-335 keV, 662 keV), and ^{237}Np - ^{233}Pa contribute to bias in the NaI assay of ^{239}Pu at 414 keV. Many of these interferences are eliminated with CZT.

The recent availability of Peltier-cooled CdTe detectors with areas larger than 1 cm^2 (thickness is limited to 3 mm by charge transport properties) has made gamma-ray isotopic measurements for uranium and plutonium truly portable. Figure 1 illustrates the good energy resolution of CdTe. Figure 3 illustrates the compact dimensions of the CdTe detector, shown in use for portable Pu isotopics measurements in a glove box. The capability of CdTe for isotopic analysis covers 3% to 30% ^{240}Pu ; it also covers ^{235}U from 0.1 to ~80%, and MOX. A 15-min. count with a CdTe detector measures the ^{240}Pu fraction to ~2% and the ^{235}U fraction to ~3%.



Fig. 3. The Peltier-cooled CdTe detector is shown measuring Pu isotopics in a glove box.

III. GENERALIZED GEOMETRY HOLDUP (GGH) ASSAY METHOD

A. Assumptions and Constraints

The Generalized Geometry Holdup (GGH) method categorizes each holdup deposit, no matter how complex, as a simple point, line, or area source.⁶ This is illustrated in Figure 4 below. The GGH assay method was developed to simplify the analysis of holdup measurements performed using NaI detectors. It can, however, be used with any detector. The analysis of holdup data using GGH requires the following constraints:

1. Radiation shielding is used on the back and sides of the crystal.
2. A cylindrical collimator is installed on the front of the crystal.
3. The detector is positioned so that the deposit can be approximated as:
 - a. a small point at its center, or
 - b. a narrow, uniform line through its center whose length exceeds its width, or
 - c. a uniform distribution that fills it (area deposit).
4. Measurements are performed at a known distance r between the detector and the deposit.

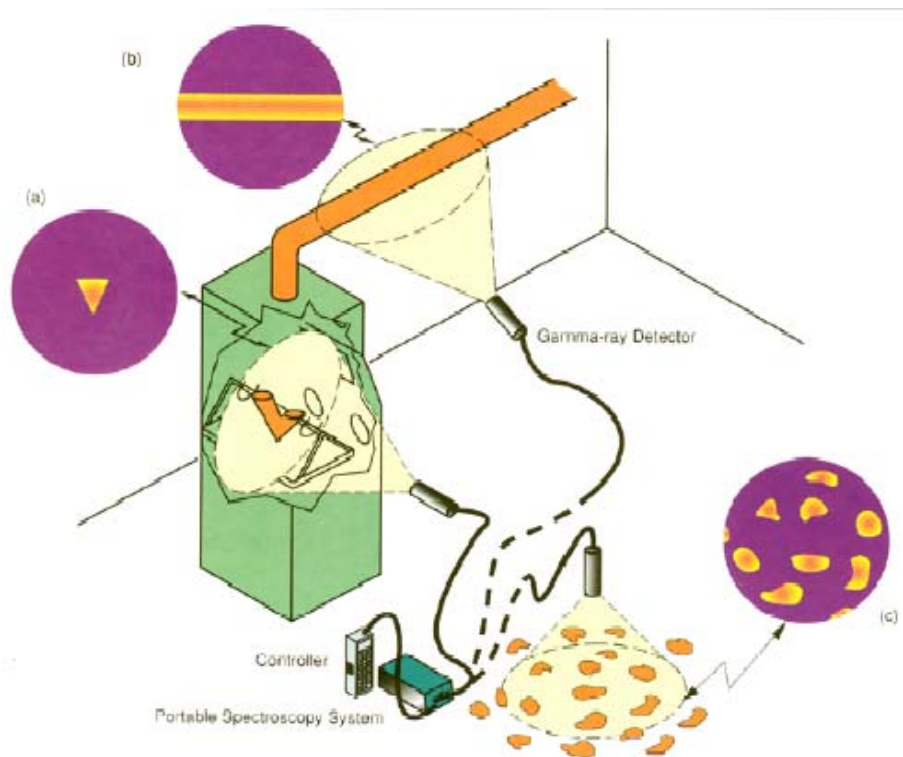


Fig. 4. Illustration of point (a), line (b), and area (c) holdup deposits.

B. Calibration

The calibration of the GGH method determines the relationship between the count rate of the measured gamma ray and the mass of the isotope of interest. Calibration for the assay of a point, line, or area deposit is accomplished with a point reference source. The response for each gamma-ray peak is measured with this source positioned on the detector axis at a known distance from the crystal. Measurements are also performed with the source displaced at fixed intervals from the crystal axis to obtain the two-dimensional radial response of the detector. These data are used to obtain the calibration for the assay of the specific isotope mass in a point, line, or area deposit. Figure 5 illustrates nine off-axis positions that can be used to determine the two-dimensional radial response of the detector. For the fixed distance, the detector response to a source at any point on the circle is the same because of rotational symmetry.

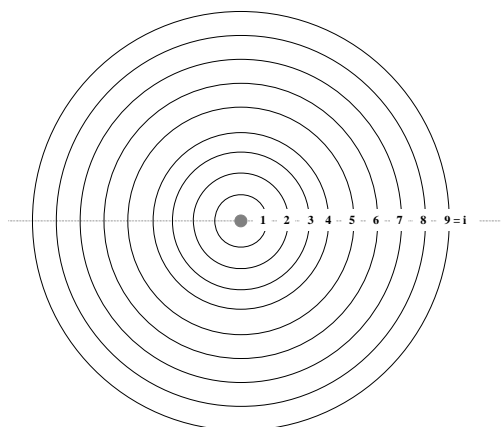


Fig. 5. Nine, equally spaced, off-axis positions of the point reference source used to measure the two-dimensional radial response of the detector.

Figure 6 shows the radial response obtained for the 414-keV gamma ray of ^{239}Pu using a 2.54-cm-diameter by 5-cm-thick NaI(Tl) detector. The length and diameter of the collimator are both 2.54 cm. The distance between the calibration source and crystal, r_0 , is 40 cm. In this example, data were obtained with the point source positioned on both sides of the axis. The data in Figure 6 are used for the calibration of the assay of the specific isotope mass in a point, line, or area deposit. The point calibration uses only the axial response C_0 (s^{-1}) because the point deposit has no finite width. The point calibration constant is

$$K_P (\text{g} \cdot \text{s} \cdot \text{cm}^{-2}) = m_0 \div (C_0 \cdot r_0^2). \quad (1)$$

The specific isotope mass measured at a distance r (C = room-background-subtracted net) is:

$$m_P (\text{g}) = K_P \cdot C \cdot r^2. \quad (2)$$

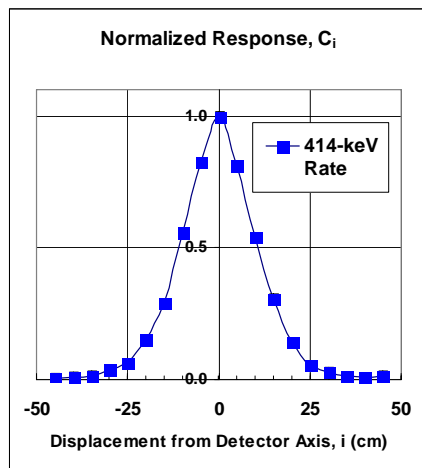


Fig. 6. The normalized count rate of the ^{239}Pu 414-keV peak vs source position. This radial response was measured at 40 cm (r_0) with a collimated NaI(Tl) detector.

For a point deposit, the specific mass is the measured mass. The random relative uncertainty, assuming only an error in C , is:

$$\sigma_R(m_P) = \sigma_R(C), \quad (3)$$

where the $\sigma_R(C)$ is the random uncertainty from counting statistics.⁷

The line calibration uses both the un-normalized axial response C_0 (s^{-1}) and a geometric parameter L that is evaluated from a sum of the normalized radial responses C_i weighted by the distance between the measurement positions.⁷⁻⁹ The line calibration constant is

$$K_L (\text{g} \cdot \text{s} \cdot \text{cm}^{-2}) = m_0 \div (L \cdot C_0 \cdot r_0). \quad (4)$$

The specific isotope mass measured at a distance r from a line deposit is:

$$m_L (\text{g/cm}) = K_L \cdot C \cdot r. \quad (5)$$

This is also called the linear density of the deposit. The random relative uncertainty in the linear density is the same as Equation 3.

The area calibration uses both the un-normalized axial response C_0 (s^{-1}) and a geometric parameter A that is evaluated from a sum of the normalized radial responses C_i weighted by the

area between the axially concentric circles that intersect the measurement positions.⁷⁻⁹ The area calibration constant is:

$$K_A (\text{g} \cdot \text{s} \cdot \text{cm}^{-2}) = m_0 \div (A \cdot C_0) \quad . \quad (6)$$

The specific isotope mass measured as an area deposit is:

$$m_A (\text{g/cm}^2) = K_A \cdot C \quad . \quad (7)$$

This is also called the areal density. The random relative uncertainty in the areal density is the same as Equation 3. By adhering to the constraints above, calibration of many geometries is accomplished with a point source.

C. Performing the GGH Measurement and Assay

Because GGH count times are often very short (5–15 s), the random uncertainty can be large for individual measurements. Propagating the uncertainties of the many measurements to get the total holdup in a facility greatly reduces the random error.

The initial assay result is the specific isotope mass for a point, line or area deposit. Three additional corrections are required for equipment attenuation, finite-source dimensions, and the self-attenuation of the deposit. These corrections are described in detail in Reference 6.

D. CORRECTION FOR EQUIPMENT ATTENUATION

The specific mass of a point, line, or area deposit, m_P , m_L , or m_A is corrected for equipment attenuation effects using the formula:

$$CF_{EQ}(Z, E\gamma) = e^{\mu \rho t}, \quad (8)$$

where ρ and t are the density and thickness of the equipment. The corrected masses are

$$m_{P,EQ} (\text{g}) = m_P \cdot CF_{EQ}(Z, E\gamma), \quad (9)$$

$$m_{L,EQ} (\text{g/cm}) = m_L \cdot CF_{EQ}(Z, E\gamma), \text{ and} \quad (10)$$

$$m_{A,EQ} (\text{g/cm}^2) = m_A \cdot CF_{EQ}(Z, E\gamma), \quad (11)$$

respectively. Because the correction is applied linearly, the relative uncertainties in $m_{P,EQ}$, $m_{L,EQ}$ or $m_{A,EQ}$ are unchanged from that in Equation 3.

If no correction for equipment attenuation is performed, the assay results will always be biased low. An estimate of the equipment attenuation based on the best information available gives a result that may be high or low for individual measurements, but the overall result tends to be much better than ignoring the correction altogether. In recent measurements of ²³⁹Pu holdup in bulk-processing equipment in glove boxes using the 414-keV gamma ray, values of $CF_{EQ}(Z, 414 \text{ keV})$ varied from a low of 1.1 (lead-lined gloves) to a high of 6.2 (steel plates on a glove-box floor).

E. CORRECTION FOR FINITE SOURCE DIMENSIONS

1. Finite Sources in Holdup Measurements

The effect of the finite dimension of a point or line deposit is substantial if the deposit width w is not small compared to the detector's field of view. The user may choose a measurement distance to minimize the finite-source effect, but in many situations this is not practical.

Figure 7(a) is an example of a holdup measurement where r is large compared to the width of the deposit in the vertical pipe. Figure 7(b) illustrates a measurement with the same detector where the diameter of the horizontal overhead duct is four times that of the pipe in Fig. 7(a).



6(a)



6(b)

Fig. 7. (a) A collimated NaI detector is positioned to measure holdup in the pipe at the location marked by the bar-code label. In this case, the finite-source effect is small. (b) The same detector is positioned at the same distance from a horizontal duct whose diameter is four times that of the pipe in (a). In this case, the finite-source effect may be large.

The correction for the finite-source dimension of a point, line, or area deposit is applied linearly to the respective specific mass, Eqs. (9), (10), or (11), that has been corrected for equipment attenuation. The correction factor for a finite-source effect is always 1 or greater. The respective specific masses corrected for finite-source effects are

$$m_{P,FIN} (g) = m_{P,EQ} \cdot CF_{FIN,P} , \quad (12)$$

$$m_{L,FIN} (g/cm) = m_{L,EQ} \cdot CF_{FIN,L} , \quad (13)$$

and

$$m_{A,FIN} (g/cm^2) = m_{A,EQ} \quad (14)$$

because

$$CF_{FIN,A} = 1. \quad (15)$$

Because the finite-source correction is applied linearly, the relative uncertainty is unchanged from that given in Equation 3.

2. Concept of a Finite Source

Figures 8-9 are sketches that illustrate finite line and point deposits.

Fig. 8. The shaded region represents a finite line deposit. It is superimposed on the field of view of the detector. The circles represent the measurement positions for the calibration.

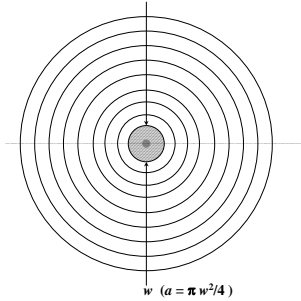
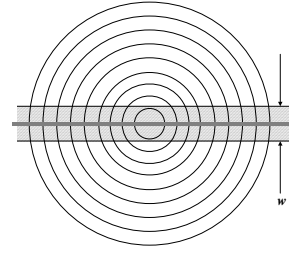


Fig. 9. The shaded area represents a finite point deposit. It is superimposed on the field of view of the detector. The finite point deposit is centered in the field of view.

3. Correcting a Measured Holdup Deposit for Finite Source Dimensions

Knowledge of w is also required for the self-attenuation corrections, as described later. It is important to perform both corrections:

- to avoid a negative bias in the measured holdup (that results from both effects), and
- to reduce the magnitude of the systematic effects associated with over-/under-estimates of the experimental width parameter w .

The six steps to apply finite-source corrections are:

1. Fit the C_i to a normalized Gaussian $G(x)$ that represents the radial response curve.

$$C(x) = G(x) = \exp[-0.5(2.354 x / \text{FWHM})^2], \quad (16)$$

where “ x ” is the source distance from the detector axis. Figure 10 is a plot of Eq. (16).

2. Measure a holdup deposit at distance r with finite dimension w .

$$w_0 = w \cdot (r/r_0)^{-1}. \quad (17)$$

3. Determine the normalized response, $C(w_0/2)$, at the outer edge ($x = w_0/2$) of the line or point from the plot (Figure 10) or from Eq. (16).

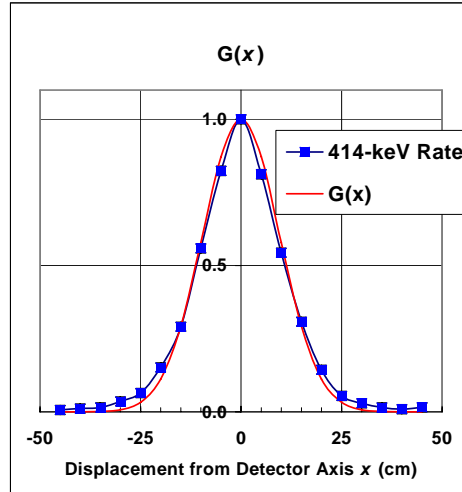


Fig. 10. The data from Fig. 6 are shown in blue. The red curve is the fit of Eq. (16) (normalized Gaussian) to these data.

4. Obtain the average of $C(w_0/2)$, the normalized response at the edge of the deposit, and 1, the normalized response at the center of the deposit, to get the effective radial response

$$C_{EFF} = [1 + C(w_0/2)] / 2. \quad (18)$$

5. Compute the finite-source correction factor for a line or point deposit

$$CF_{FIN, L} = (C_{EFF})^{-1}, \quad (19)$$

$$CF_{FIN, P} = (C_{EFF})^{-1}. \quad (20)$$

6. Apply the appropriate point or line finite-source correction to the equipment-attenuation corrected specific mass, Eq. (12) or (13).

In recent GGH measurements of plutonium holdup¹⁰ in high-throughput, bulk-processing equipment inside of glove boxes, the values of $CF_{FIN, L}$ obtained varied from 1 (for area deposits on glove-box surfaces) to 1.25 (for line deposits of powder accumulated in troughs on the glove-box floor). The procedure described above applies to holdup deposits measured with a cylindrically collimated gamma-ray detector of any type (NaI, BGO, HPGe, CdZnTe, etc.) and any gamma-ray energy.

F. CORRECTION FOR SELF-ATTENUATION EFFECTS

1. Self-Attenuation Effects in Holdup Measurements

A characteristic of special nuclear materials is the self-absorption of gamma rays emitted by the material. The corrections are important for any gamma-ray measurement. The magnitude of the self-attenuation can be determined if the areal density of the deposit (corrected for room background, equipment attenuation, and finite-source effects) is known. Because the atomic numbers (Z) of actinides are large and the actinide density dominates the density of the deposit,

the actinide areal density is sufficient to perform the correction. The key to this correction is that the specific mass of the actinide obtained from the holdup measurement can be converted to the uncorrected areal density of the actinide. The holdup measurement of a point deposit gives the uncorrected mass of the actinide isotope. This can be converted to an uncorrected areal density by dividing by the area a of the point deposit, which is

$$a = \pi \cdot (w/2)^2. \quad (21)$$

2. Determining the Self-Attenuation Correction from the Measured Areal Density

The areal density of a deposit is its mass per unit area (g/cm^2). The measured specific mass of a deposit can be used to obtain the actinide areal density, $(\rho x)_{\text{MEAS}}$. The true areal density, (ρx) , is obtained using this equation:

$$(\rho x)/(\rho x)_{\text{MEAS}} = \mu(\rho x)/[1 - e^{-\mu(\rho x)}], \quad (22)$$

where μ is the mass attenuation coefficient (cm^2/g) of the element, compound, or mixture at the energy used for the measurement.¹¹ Rearranging Eq. (22) gives the true areal density (corrected for self-attenuation) as a function of the measured:

$$(\rho x) = -(\ln[1 - \mu(\rho x)_{\text{MEAS}}])/\mu. \quad (23)$$

Table 3 lists the μ values for three common forms of uranium deposits (U metal, UO_2 and U_3O_8) and two common plutonium materials.

Table 3.
Gamma-ray Mass Attenuation Coefficients

Material	E_γ (keV)	μ (cm^2/g)
U	186	1.46
UO_2	186	1.30
U_3O_8	186	1.26
Pu	414	0.270
PuO_2	414	0.250

3. Correcting a Measured Holdup Deposit for Self-Attenuation

An over- or underestimate of w leads to an under- or over-correction for self-attenuation. This is opposite the case of the finite-source correction. Thus, for a limited range of deposit thickness, the effects of errors in the estimates of w tend to compensate when both corrections are performed. Unlike the other corrections, the correction for self-attenuation, Eq. (22), is a nonlinear function of the specific mass of the deposit and it must be applied after the other corrections. For the same reason, and because the self-attenuation is determined from the areal density of the deposit, the isotope fraction must be used to convert the measured areal density of the isotope to that of the element. If more than one actinide is present, both the element and isotope fractions are multiplied for use in the conversion step.

For measurements of ^{239}Pu in bulk-processing equipment inside glove boxes using the 414-keV gamma ray, the magnitude of the correction for self-attenuation was as large as 1.11 for powder deposits on the glove-box floor.

The procedure for self-attenuation corrections is currently automated¹² in the stand-alone VisualBasic program *SelfAttn v. 1.0*. The automation is possible because of the simplicity of the approach. This software module is incorporated into the HMS3 software, which automates the plant-wide portable measurement and tracking of holdup.^{13, 14}

IV. HOLDUP MEASUREMENT SYSTEM EXAMPLES

The Integrated Holdup Measurement System at the Y-12 HEU plant in Oak Ridge, Tennessee, is a good example of a comprehensive holdup measurement system.¹⁵ The Y-12 facility identified many thousands of measurement points, each marked with a bar code label. Operators carry a small multichannel analyzer (MCA), a collimated NaI detector, and a handheld bar code reader with a data logger/controller. Thousands of locations are measured each month. Data from the data logger are downloaded into a computer running a program called HMS4. This has been used successfully for more than seven years. An extensive study was made of system performance using simulated holdup situations such as pipes, ducts, and V-blenders with known U or Pu sources. Figure 11 below shows a technician at Y-12 measuring an overhead duct.



Fig.11. GGH applied in a uranium facility to measure an overhead duct.

Figure 12 shows a similar measurement system in use at a plutonium processing facility. In this case a telescoping pole, such as used by house painters, is used to position the NaI detector near the overhead pipes and ducts. Figures 13 and 14 show other ^{235}U holdup measurements in a uranium processing facility using Ge, CZT, and NaI detectors. Figure 13 shows a very large overhead duct being measured with a portable Ge detector weighing ~10 kg with collimator. Figure 14 shows CZT and NaI detectors weighing ~1 kg each with collimators. The greater portability of the room-temperature detectors is essential for most holdup measurements.



Fig. 12. A compact NaI detector is shown during measurements of plutonium deposits in overhead ducts.



Fig. 13. A large overhead duct is measured from below with a collimated Ge detector.



Fig. 14. Measurements of ^{235}U deposits in a filter system performed with CZT and NaI.

The Rocky Flats Plant near Denver, Colorado, was closed in 1989, dismantled, cleaned, and converted into an environmental park. During its operating lifetime (~40 y), Rocky Flats accumulated large quantities of plutonium holdup in the gloveboxes, filters, calciners, pipes, and air ducts of several major processing buildings. This holdup was a significant health and criticality safety concern, and at times was a major contributor to the MUF for the facility.

During the decommissioning of the processing buildings, the holdup measurement campaigns were among the largest and most extensive ever reported. The holdup measurement teams pioneered the use of medium-resolution BGO detectors, and the use of measurements made with the detectors in contact with pipes or ducts. Although this approach is more susceptible to uncertainties in material distribution than the GGH methodology, it allows routine measurements to be made more quickly. As buildings were decommissioned and the process lines were removed and cleaned out, it was often possible to obtain comparisons between the measured holdup and cleanout values. The overall results of numerous measurements of extended equipment lines tended to be unbiased.¹⁶

V. NEUTRON HOLDUP MEASUREMENTS

Nuclear material processing equipment can be massive and extensive. High-throughput facilities may contain multi-kilogram deposits. The high attenuation of such equipment and deposits may challenge the capability of gamma-ray holdup measurements. In such cases, the high penetrability of neutrons may offer a more reliable option. Neutrons can be detected from pumps, furnaces, and other heavy equipment that are too dense to permit gamma rays to escape. It is difficult to shield neutron detectors from room background, but counting coincident neutrons from spontaneous fission can effectively reduce background. Large polyethylene-moderated, ^3He slab detectors have been used successfully to quantify in-process plutonium in glove boxes. Although the spontaneous-fission neutron yield from uranium is low for coincidence counting, the high α, n yield from fluorine enables measurements of uranium deposits using total neutrons from UF_6 and UO_2F_2 in enrichment plants.

Light weight, directional, portable neutron counters are difficult to design because of the need for a polyethylene moderator surrounding the ^3He tubes. However, several reasonably portable detectors have been designed and used for holdup measurements. The original portable counter was the Shielded Neutron Assay Probe (SNAP-II) fabricated in 1975 (see Figure 15). The SNAP-II had two 20-cm-long ^3He tubes in a 12.7-cm-diam polyethylene cylinder, wrapped in cadmium and surrounded for 240° by a 5.7-cm-thick directional shield. The intrinsic efficiency of the SNAP-II for fission neutrons was $\sim 17\%$ and it weighed ~ 10 kg. The SNAP-II was used to measure uranium holdup in operating and shut down gaseous diffusion enrichment plants and plutonium holdup in several scrap recovery facilities.



Fig. 15. Shielded Neutron Assay Probe (SNAP-II) used for U and Pu holdup measurements in heavily shielded situations.

The Portable Handheld Neutron Counter (PHNC) shown in Figure 16 is a newer counter with four ^3He tubes. The PHNC has no directionality, but it is more efficient and weighs less than the SNAP-II. The PHNC is designed for either singles counting of wide-area holdup sources or coincidence counting, with two PHNC slabs, of small containers of Pu materials.¹⁷

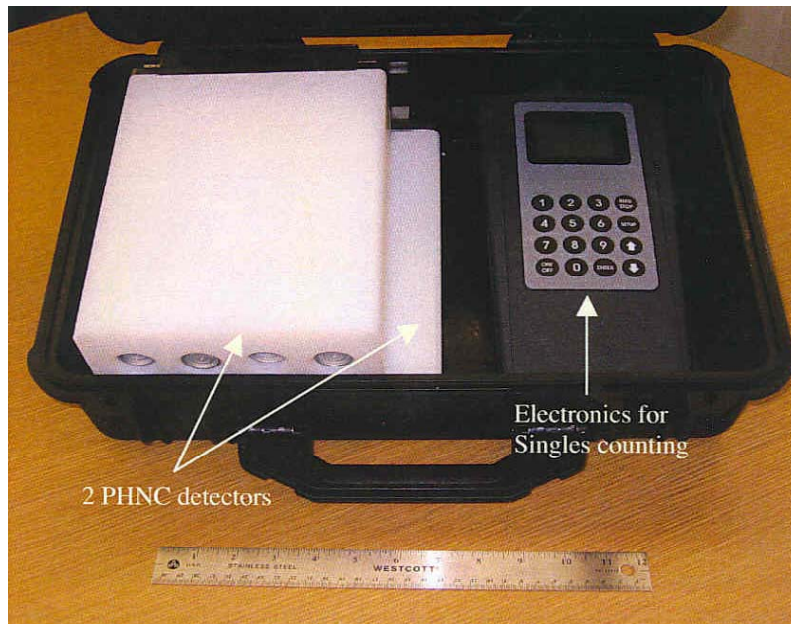


Fig. 16. Two Portable Handheld Neutron Counters (PHNC) and a related electronics package.

Large slab detectors can provide higher efficiency and better directionality if heavy shielding and collimation are added. Such detectors are too heavy for handheld operation, but they can be moved with carts or can be permanently installed to measure in-process inventory or holdup after cleanup. The slab detectors shown in Figure 17 were designed to measure holdup and in-process inventory at uranium enrichment plants.



Fig. 17. Wide-area neutron detectors used to measure holdup at uranium enrichment plants.

A new neutron holdup assay method for enrichment facilities, or in any facility with a large distributed volume of material, was recently described.¹⁸ The Distributed Source-Term Analysis

(DSTA) technique uses Monte Carlo modeling of a centrifuge enrichment cascade hall to derive a calibration curve relating the average neutron count rate to the mass of uranium holdup. Then a portable counter, similar to the PHNC, is used to survey the average neutron count rate in the hall. This approach avoids the high attenuation problems of gamma-ray measurement, the difficulties in measuring individual pieces of equipment to obtain the total holdup, and the long measurements required to assay the entire process line.

Large slab detectors have also been used to measure plutonium holdup in rotary calciners, hydrofluorinators, and other large, highly attenuating items. Two slab detectors, each 50-cm tall by 100-cm-wide by 8-cm-thick polyethylene with ten 90-cm-long ^3He tubes, were placed in a rack and moved to various locations around a rotary calciner used to dry plutonium peroxide. The calciner was inside a glove box. The quantity of holdup was sufficiently high to use neutron coincidence counting, so the measurements were less sensitive to background neutrons. The detectors were calibrated with a 2-kg PuO_2 standard. After one of the measurement campaigns, the calciner was cleaned out and the recovered plutonium measured. The holdup assay agreed very well with the recovery value.¹⁹

Large neutron slab detectors have been used in the Plutonium Fuel Production Facility (PFPP) in Tokai-Mura, Japan, to measure holdup in glove boxes inside this automated MOX fuel fabrication facility.²⁰ These Glove Box Assay Systems (GBAS) were 160 cm high, 100 cm long, and 7.6 cm wide (see Figure 18). Each slab contained twenty 152-cm ^3He tubes. Monte Carlo calculations were used to design the detector and study its response before installation. Six slabs were originally installed in pairs on either side of a glove box. The slabs could be moved remotely to measure different locations on a glove box. A standard matrix of measurement positions was assigned for each glove box and software written to collect, analyze, and combine all the measurements. Measurement data from this system are shared by the IAEA and the facility operator. Experience at PFPP has shown a measurement uncertainty of ~5% for neutron assay and 25-30% for gamma-ray assay.

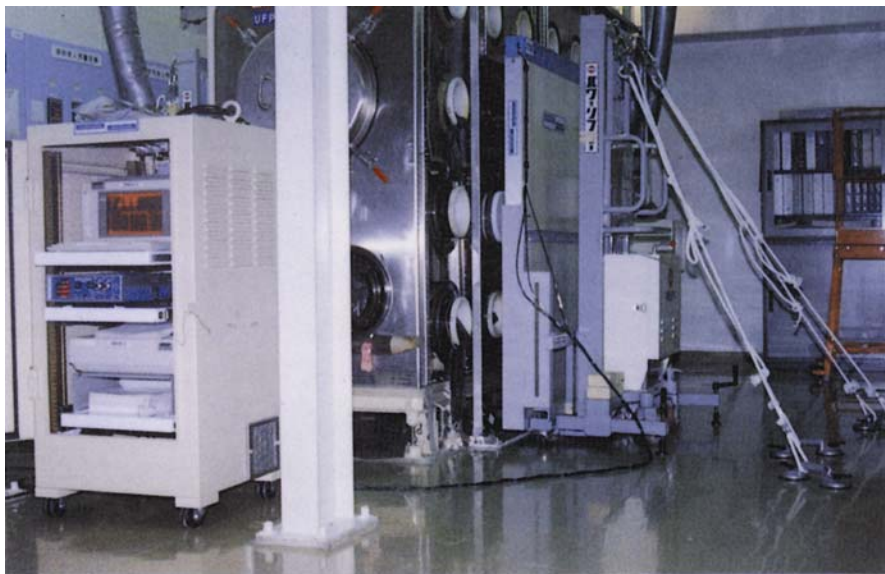


Fig. 18. Glove Box Assay System (GBAS) installed at PFPP in Tokai-mura, Japan.

VI. ACCURACY OF HOLDUP MEASUREMENTS

The precision or random error can be readily determined for all NDA measurements including holdup. Because of the many measurements performed, the overall precision is usually of the order of a few percent or less. However, the accuracy or systematic error is very difficult to determine, because it is difficult to know the true mass of nuclear material held up in the equipment of a complex facility. Often, the accuracy estimate for a holdup campaign is simply the "best guess" of the operator based on judgment and experience. Such estimates are typically in the range 25 - 50% or more, because of the many unknown factors and assumptions required to calculate the nuclear material mass. In some cases, e.g. glove boxes, known standards can be introduced and measured in addition to the holdup. In a few cases, an effort was made to clean out and recover the measured material which was then analyzed destructively and compared with the measured holdup. A complete clean out is usually difficult and costly, but this is the best way to determine holdup assay accuracy.

In the early 1980s, a holdup measurement campaign was conducted at a shut down part of the Portsmouth Gaseous Diffusion Plant in Piketon, Ohio. Gamma-ray measurements were made with a collimated NaI detector and neutron measurements were made using the slab detectors shown in Figure 17. A total of approximately 250 stages (converter, cooler, compressor, and piping) were measured during the campaign. Afterwards, three cells (12 stages each) were cleaned out and the uranium recovered. The U was also measured and recovered from an isolated converter. The results from this are summarized in Table 4. Because the gamma-ray measurements only covered the converters, they should only be compared with the neutron assay of the isolated converter. These results are typical of what one finds in such holdup studies.²¹

A six-year study was conducted on the accuracy and precision of holdup measurements using the GGH (gamma ray assay) approach to measure simulated holdup situations with well known nuclear material standards. A series of simulated holdup sources were fabricated for this study and a holdup training course; they included a pipe array, a steel pipe, an aluminum pipe, a rectangular ventilation duct, a V-Blender, and a contaminated spot on a floor. These were "salted" with U or Pu fuel rods, U metal foils, and small cans of UO₂ or PuO₂. Table 5 summarizes the results of this study which included measurements made by many people from students to holdup experts. The results shown here are "best case" vis-à-vis holdup assay accuracy.²²

Table 4. Evaluation of PGDP Holdup Assay

Cell	n kg U ^a	γ kg U ^b	Recovery kg U
A	177	45	120
B	32	3	28
C	29	12	25
isolated converter	9	10	7

- The neutron counters were not well collimated and measured an entire stage and double-counted the cooler.
- Gamma-ray measurements covered only the converters.

Table 5. GGH Holdup Assay Evaluation

	$^{235}\text{U}^{\text{a}}$	$^{239}\text{Pu}^{\text{a}}$
pipe array	0.90	0.72
V-blender	1.22	1.02
Al pipe	1.03	0.97
Steel pipe	0.97	1.47
floor spot	0.96	n/a
duct	1.07	0.96

a. Number listed is the average ratio of measured U or Pu to the reference value.

The Rocky Flats Environmental Technology Site (RFETS), located near Denver, contained 802 buildings. The plant, which manufactured plutonium parts for nuclear weapons, was closed in 1989 and subjected to a 10-year cleanup campaign that ended in 2005 when RFETS was turned into a national wildlife refuge. During this period, $3.5 \times 10^5 \text{ m}^2$ of buildings were dismantled and over 220 kg of plutonium holdup measured by a staff of 15. Holdup measurements included nearly 7 km of ductwork ($\sim 3 \text{ gPu/m}$), 1497 gloveboxes, and over 300 plutonium process tanks. Gamma-ray measurements were performed using HPGe and Bismuth Germanate detectors and the GGH procedures (see Figure 19). All of the measured equipment was cleaned out and the recovery values can be compared with the pre- and post-cleanout NDA holdup measurements. The cleanout data were generally within 20% of the measured holdup. Some specific building values are listed in Table 6.²³

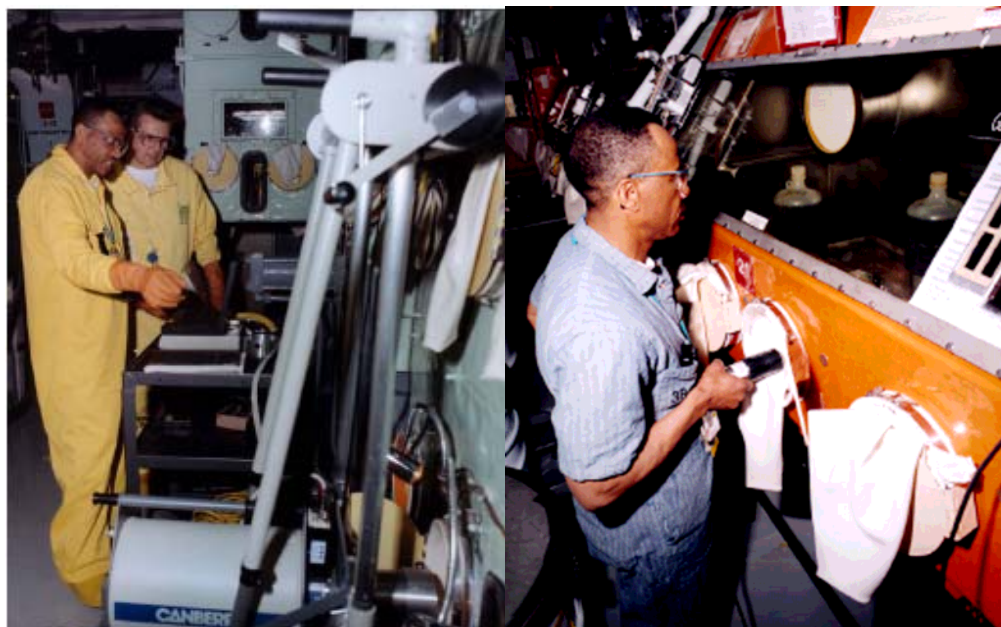


Fig. 19. Holdup measurements at RFETS: Ge on left, BGO on right.

Table 6. RFETS Holdup Data

Building	Measured/Recovered Pu
B-371 Gloveboxes	1.09
B-307 Ductwork	1.06
B-779 Total Holdup	1.13
B-A Total Holdup	1.17
B-B Total Holdup	0.97
B-C Total Holdup	1.04
B-D Total Holdup	1.03

VII. SUMMARY

A generally useful approach to the quantitative measurements of holdup invokes simple, geometric models (point, line, and area, GGH). The result is that simple calibration procedures and assay algorithms apply to most deposits. Corrections are applied for room background, equipment attenuation, the finite dimensions of point and line deposits, and the self-attenuation of gamma rays by the deposit. Knowledge of the width of a point or a line deposit is sufficient to perform the finite-source and self-attenuation corrections. The error in the finite-source correction from uncertain knowledge of the width parameter is partially cancelled by an error of the opposite sign in the self-attenuation correction. For area deposits, only the self-attenuation correction applies. It is independent of an additional parameter and is determined directly from the uncorrected holdup result.

There is a tendency for the relative finite-source effects to be larger for holdup measurements of ^{239}Pu and for the self-attenuation effects to be larger for holdup measurements of ^{235}U . This is a result of the gamma-ray assay energy. The higher energy gamma ray from ^{239}Pu penetrates the detector shield readily, causing users to measure closer to the deposits. A relative enlargement of the finite source dimension is the result of the reduction in the detector's field of view at the smaller measurement distance. The lower energy gamma ray used to measure ^{235}U is more affected by self-attenuation. However, the detector shielding is quite effective at eliminating room background, causing users to perform measurements at greater distance from the deposits. The result is a reduction in the relative finite source dimension.

A major source of bias arises from the relatively poor resolution of the portable, compact scintillator detectors that are used for holdup measurements. Effects of interfering gamma rays from complex spectra can contribute to a positive or negative bias in the holdup result obtained from measurements with scintillators. The portability of a room-temperature detector cannot be sacrificed in holdup applications. Therefore, recent progress in the manufacturing of large, room-temperature, solid-state gamma-ray detectors that perform better than scintillators is a welcome advance in technology for improved accuracy in holdup measurements.

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