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Evaluation of High-Resolution Gamma-Ray Methods for Determination of Solid Plutonium Holdup in High-Throughput Bulk-Processing Equipment

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EVALUATION OF HIGH-RESOLUTION GAMMA-RAY METHODS FOR DETERMINATION OF SOLID PLUTONIUM HOLDUP IN HIGH-THROUGHPUT BULK-PROCESSING EQUIPMENT

by

P. A. Russo, R. Siebelist, J. A. Painter, and J. E. Gilmer

ABSTRACT

The first field testing of high-resolution gamma-ray methods and current hardware and software technologies applied to measurements of solid plutonium holdup in high-throughput bulk-processing equipment has shown promising results. The gamma-ray assays agree reasonably with the quantities recovered from the cleanout of a continuous-feed rotary calciner. The limitations on these measurements are primarily the uncertainties in the equipment attenuation factors. Approaches to reduce these uncertainties and to correct for effects of deposit self-attenuation are discussed. The criteria for success in measurements of this type are reviewed, and the impact of anticipated holdup on the recommended startup procedures for new facilities is addressed.

I. INTRODUCTION

To increase productivity, DOE facility operators seek to eliminate the need for the time-consuming disassembly and cleanout of bulk-processing equipment to determine the holdup component of the special nuclear material inventory. On-line nondestructive assay of holdup is a potential answer. The rotary calciner in Building 771 at the Rocky Flats plant provides an example of such a holdup measurement need. Past efforts to use polyethylene-moderated ^{3}He slab detectors placed at line for counting coincident neutrons originating from calciner holdup deposits proved successful only when the deposits were so large that a cleanout was mandated anyway.[1] The uncertainty in the neutron assay of smaller deposits (several kilograms) was boosted by the uncertainty in the (relatively increased) background count rate and by the still large accidental coincidence rate. In this case, a practical approach to merely verifying the holdup quantity using neutron methods was difficult to conceive.

The use of gamma-ray measurements in these applications offers the advantages of improved signal-to-background ratios, the ability to localize deposits, and the ability to establish a clearer distinction between the signals from the holdup deposits and background signals. However, and unlike the situation for neutron assays, large holdup deposits in bulk-processing equipment can result in very large gamma-ray self-attenuation and equipment attenuation factors. The high-resolution gamma-ray approach introduces the possibility of using multiple-energy gamma-ray assays and transmission measurements to help diagnose, reduce, and perhaps eliminate biases from attenuation effects. Furthermore, current technologies in high-purity germanium (HPGe) detectors, portable personal computers, and new software for automating holdup measurements now make practical the multiple-energy HPGe measurements in portable holdup applications. Field experience has been gained recently in HPGe measurements of the Rocky Flats calciner during a shutdown for inventory.

Applying a generalized-geometry assay approach to the numerous and varied holdup measurement problems in a production facility is a very important step in reducing the "uniqueness" of each holdup measurement. This is essential to achieving a practical plan for plant-wide holdup measurements in the DOE facilities. The generalized-geometry approach has been used in the calibration and measurement of the calciner holdup.

II. EQUIPMENT AND SOFTWARE

The hardware used for the holdup measurements at Rocky Flats is pictured in Fig. 1. A commercial photographic stand with a tripod wheel base and a

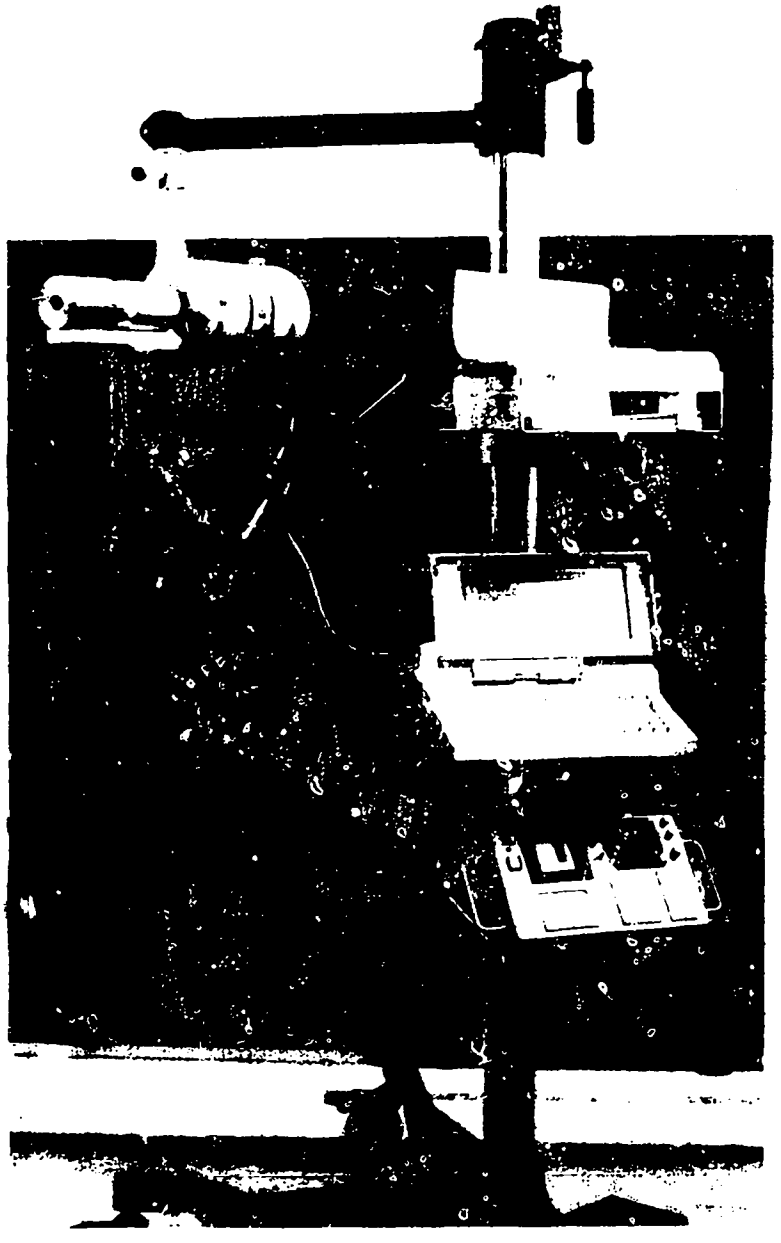

Fig. 1. Hardware for the portable holdup measurements. The shielded HPGe detector is mounted on the counterweighted arm of the movable stand (top). Below it are the printer, personal computer, and portable multichannel analyzer, each on a movable shelf. All units can operate with battery power.

counterweighted arm capable of more than 2 m of vertical travel supports the detector, portable multichannel analyzer (PMCA), laptop personal computer, and printer. The PMCA, personal computer, and printer are mounted on separate, movable, custom-designed trays.

Within the last several years, portable HPGe detectors have demonstrated improved performance and reliability in laboratory and field applications. A recently available commercial, custom designed HPGe detector features a significantly reduced total weight (for the collimated shielded detector) despite substantial shielding requirements for plutonium holdup measurements. The detector includes both a 2.5-cm-thick tungsten backshield inside the cryostat directly behind the coaxial HPGe crystal and a stepped-down end cap diameter (from 7 cm to 4 cm) at the crystal location, minimizing the weight of the external tungsten shield. Figure 2 shows the detector and the external shield, which provides a 1.6-cm thickness of tungsten around the crystal with a 2-cm-diam by 2.5-cm-deep cylindrical collimator. The total mass of this equipment

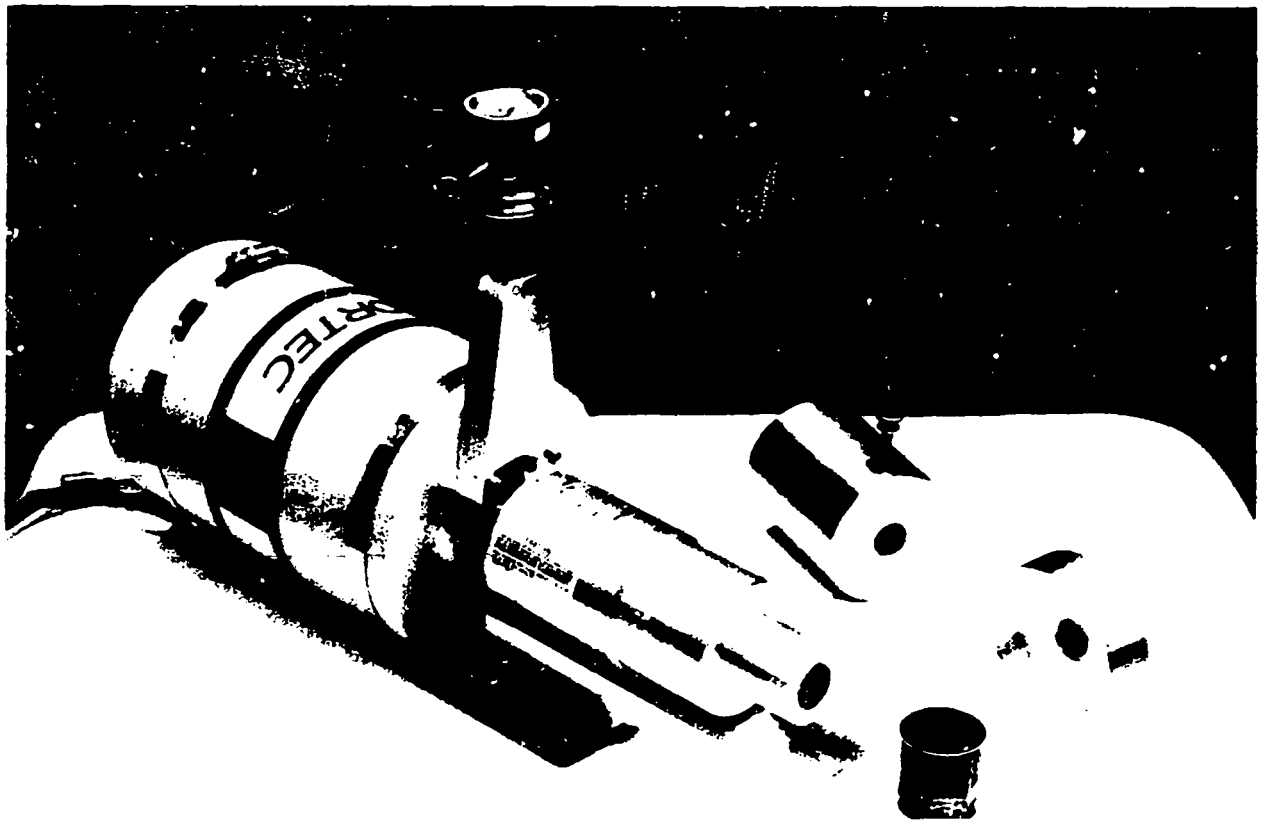

Fig. 2. Custom-designed commercial HPGe detector (left) is shown with cadmium end cap (front, center), external tungsten shield (back, center) and external source (right). Detector is shown fitted with hardware for mounting purposes.

(including the external shield) with a full (24-h holding capacity) liquid nitrogen dewar is 11.3 kg; the mass of the external shield is 4.5 kg. Previous in-plant experience in plutonium holdup measurements[2] has demonstrated the need for the 1.6-cm thickness of tungsten, and yet the total weight of this shielded detector (roughly that of a standard lead brick) makes possible a rapid sequence of measurements at different arbitrary locations in the plant. This shielded detector is shown in Fig. 1 mounted on the counter weighted arm of the movable holdup equipment stand. The mounting device includes a ball pivot for flexibility of detector positioning.

Because reducing and analyzing the data from multiple-energy gamma-ray spectra is complex, automation is essential in using HPGe detectors for multiple-energy holdup assay. A prototype software package, developed for field use in portable gamma-ray holdup assays, was used to automate the Rocky Flats calciner holdup measurements. The software controls the PMCA to automatically acquire, reduce, and analyze the gamma-ray spectral data. Use of the code in on-line applications requires an IBM-PC-compatible computer, a PMCA, a printer, and a cylindrically collimated [HPGe or NaI(Tl)] gamma-ray detector. Off-line, the PMCA gamma-ray spectral data stored on floppy diskette or PMCA data tape can be analyzed by the software.

The software tests the quality of each gamma-ray spectrum by evaluating deviations from expected photopeak centroids, photopeak widths, and count rates for each acquisition. The photopeak and background regions of interest (ROIs) can be defined either by channel number or by energy. The ROIs defined by energy are set according to a unique energy calibration for each spectrum based on the measured centroids of designated photopeaks, thus eliminating concerns about long-term drifts in electronic gain.

The code accesses a designated file of parameters (from multiple stored parameter files) that defines the PMCA setup values, ROIs, photopeak identifications, photopeak and spectrum quality requirements, measurement control criteria, calibration constants, attenuation factors, calibration geometries, and default variables associated with the data acquisition cycles. The availability of multiple parameter files enables (1) the equipment to be used with various detectors and collimation geometries, (2) the assays to be performed for various isotopes, and (3) the measurements to be applied to various sets of process equipment, simply by assigning the (new) appropriate parameter file

before running the software. The automated hardware setup at startup or restart of the software requires about 1 min, after which data acquisition can begin.

In the background option, the software acquires and reduces a background spectrum. The net count rate results for the photopeak energy region are stored for subtraction as background from count rates obtained from all subsequent spectra. Precharacterized standards are measured in the accuracy check option, and the results are compared to stored values. In the assay option, the code reduces the spectral data and applies the appropriate calibration to give quantitative results for holdup. In all options, multiple gamma- ray peaks can be analyzed.

The assay option requires the operator to define the holdup source geometry ("point," "line," or "area") in the detector field of view and the source-to-detector distance for other than area sources. The subsequent assay results are computed in units of g, g/cm, or g/cm^2 for the three geometries (respectively) where the mass is the isotope mass. When known, the effects of equipment attenuation can be included. The multiple-energy assay corrected for equipment attenuation flags self-attenuation effects when deposits are large. A separate (transmission) measurement should be performed to measure these effects. Otherwise, the flagged assay result at the highest photopeak energy is a lower limit on the actual holdup. In a separate option, the software also automates a point source calibration procedure to determine and store the calibration constants for each of the three holdup geometries. With multiple assay photopeaks, the complexity of the calibration requires that this procedure be automated. The calibration and assay formalism used in this software has been described previously.[2]

III. OVERVIEW OF METHOD

Figure 3 shows the rotary calciner at Rocky Flats labeled in a block diagram of a layout of the process area. Wet plutonium peroxide from the precipitation process is fed into the 15-cm-diam, 230-cm-long rotating calciner tube by the 6-cm-diam auger drive, which extends 54 cm into the outer calciner tube. A slight downward tilt causes the feed material to tumble downstream toward the product hopper. Three steel tumbler rods approaching 2 m in length

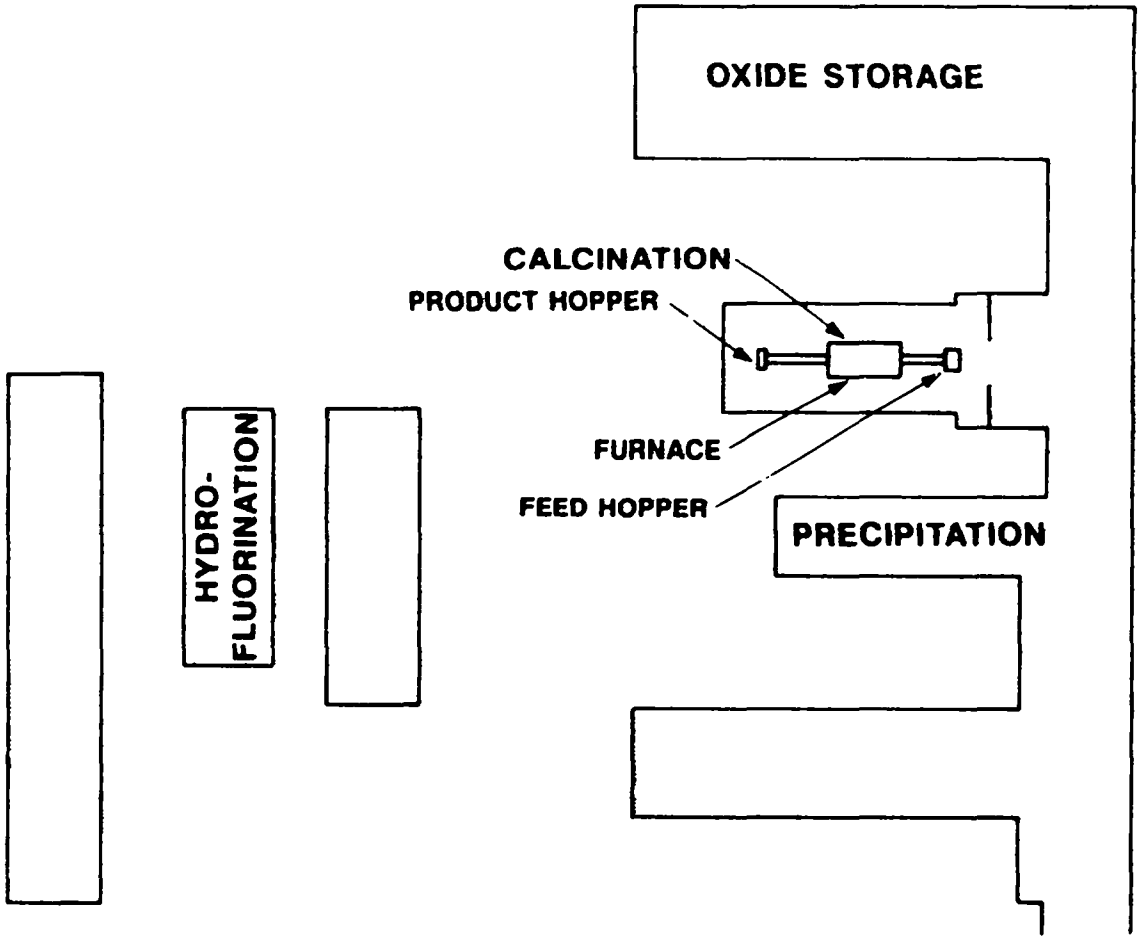

Fig. 3. Approximate process layout in the calciner area. The calciner gamma-ray holdup measurements were performed on the maintenance side (closest to the precipitation line) of the calciner glove box. The location of the hydrofluorinator (nearly identical to the calciner in design) is also shown.

prevent caking of peroxide/oxide within the calciner tube. The wet peroxide dries as it passes through the furnace section. At the product end of the calciner, the product material falls from the calciner tube into a hopper, accumulates in weighed amounts in cans. The product, PuO_2, feeds the hydrofluorination process, also labeled in Fig. 3.

The wet peroxide emerges from the auger (entering the calciner tube) approximately 50 cm downstream from the feed end of the calciner tube. However, feed material has been known to move and lodge upstream of this entry point causing calciner holdup accumulations to be larger at these locations. The lower portion of the bellows has been particularly susceptible to this problem. To a significantly smaller degree, the bellows at the product end has had a similar problem. Gross or loose accumulations within the tube can often be removed relatively easily (without a total calciner disassembly), but material

that lodges in the bellows can only be dislodged and removed by completely disassembling the calciner.

Figure 4 shows the calciner assembly, with a superimposed drawing of the viewing and glove ports on the maintenance side of the calciner glove box. The HPGe detector was positioned at two selected glove ports, shaded with concentric circles, for background measurements, and at the five glove ports shaded with the square grids for the calciner assay measurements. The detector fields of view for these measurement locations are shown schematically in Fig. 5, which is a cross section of the calciner glove box (perpendicular to the calciner axis) viewed from the product end of the calciner.

The fields of view in Fig. 5 show that the calciner assays from four of the five assay glove ports are susceptible to biases that would result from accumulations of plutonium on the bottom of the glove box. This emphasizes the need for cleaning out the glove box before the holdup measurements are performed.

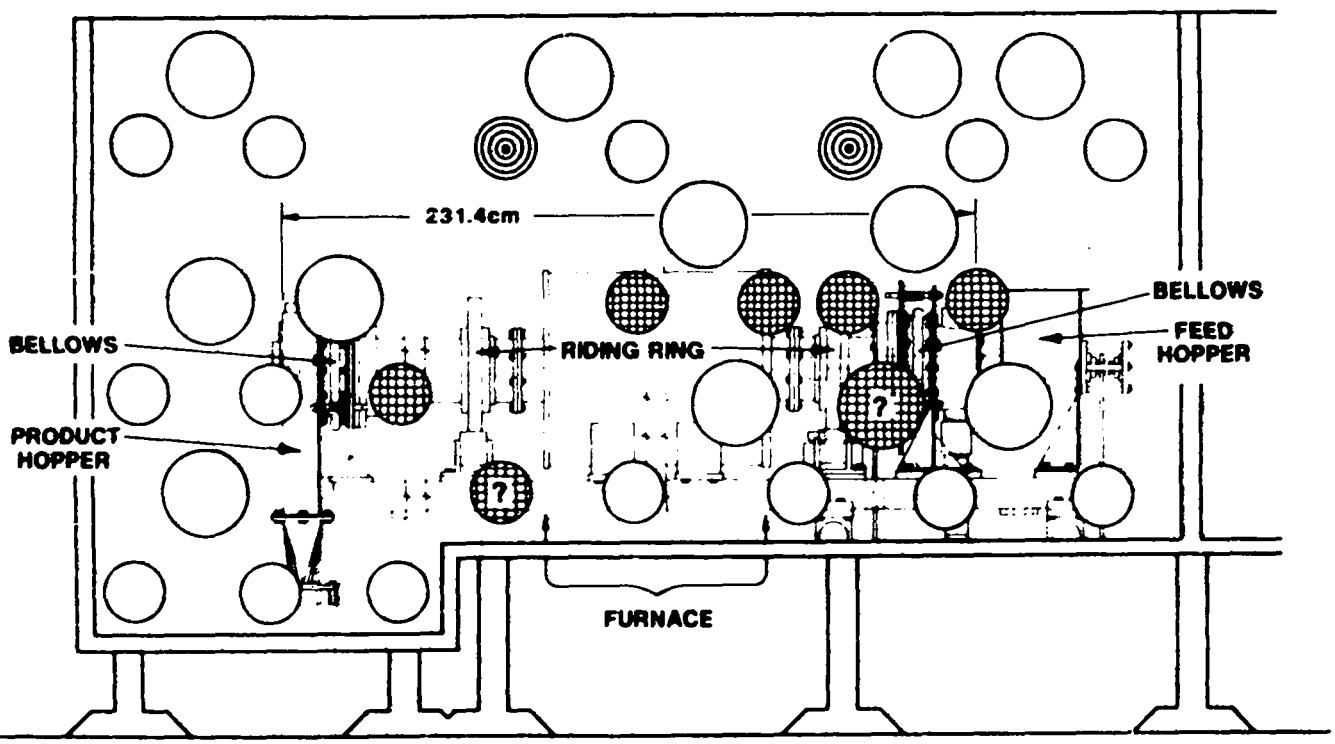

Fig. 4. Calciner assembly with (superimposed) glove ports (smaller diameter circles) and viewing ports (larger diameter circles) projected from the maintenance side of the glove box. The square grid and concentric circle shading corresponds to assay and background measurements ports. The ? labels indicate potential (untested) assay ports.

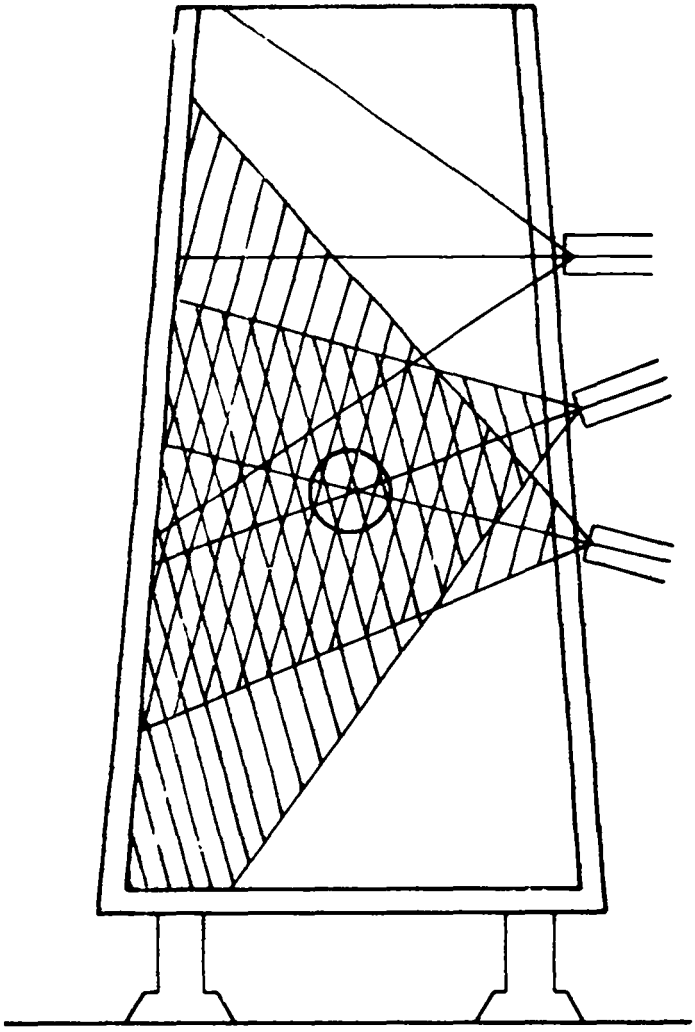

Fig. 5. Detector fields of view (looking from the calciner product end) for the background (upper position) and assay measurement locations.

The fields of view in Fig. 5 also demonstrate that the calciner tube diameter is small relative to the diameter of the detector field of view at the calciner location, justifying use of the line-source approximation for uniform deposits along the calciner axis. This is further emphasized by Fig. 6, which shows only a small decrease in the relative detection efficiency with vertical displacement of source material above and below the center of the calciner axis within the vertical dimensions of the calciner tube.

The use of the line-source approximation also requires that the uniformity of the deposit be verified, or that the entire calciner length be appropriately "sampled" in the viewing process during the measurements. The actual "sampling" achieved in the measurements in this case is shown in Fig. 7, which depicts the measurement glove ports and the full fields of view on the calciner axis superimposed on a sketch of the calciner tube. Except for a gap near the product end (left extreme in Fig. 7) of the calciner, the optimum overlap (at the half maxima of radial counting efficiency profiles for adjacent fields of

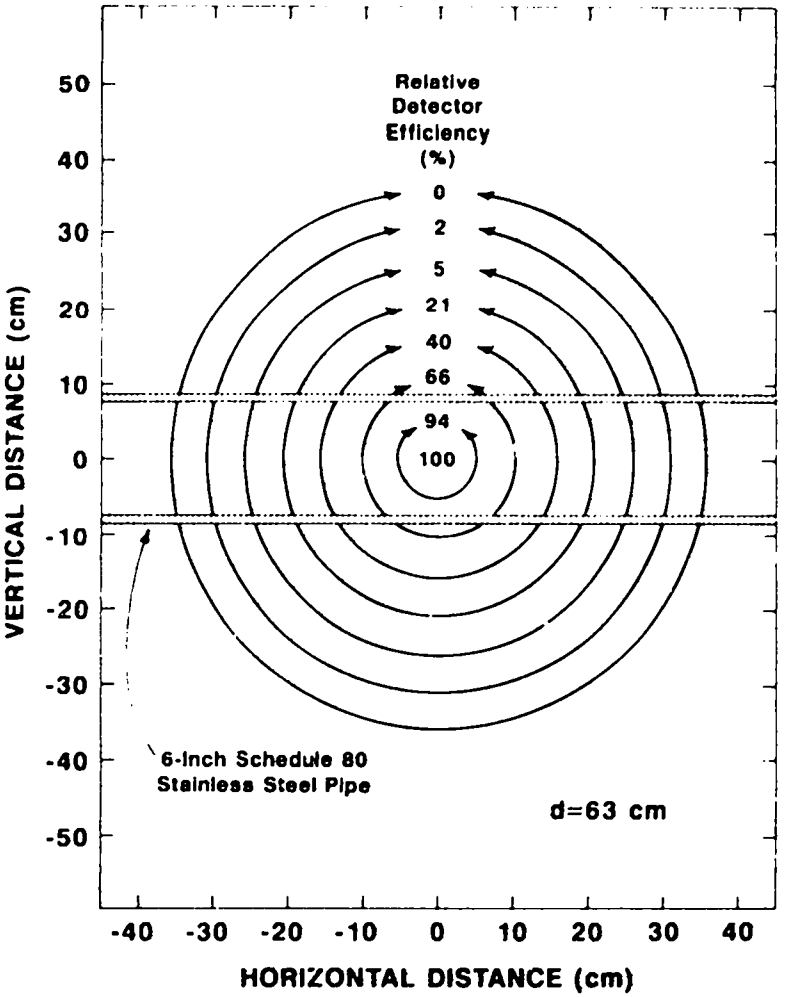

Fig. 6. Constant-detection-efficiency contours (projected normal to detector axis) on the calciner axis corresponding to the geometry for the holdup assay. The calciner tube is also outlined to the same scale. The calciner (center) to crystal distance, d, is 63 cm.

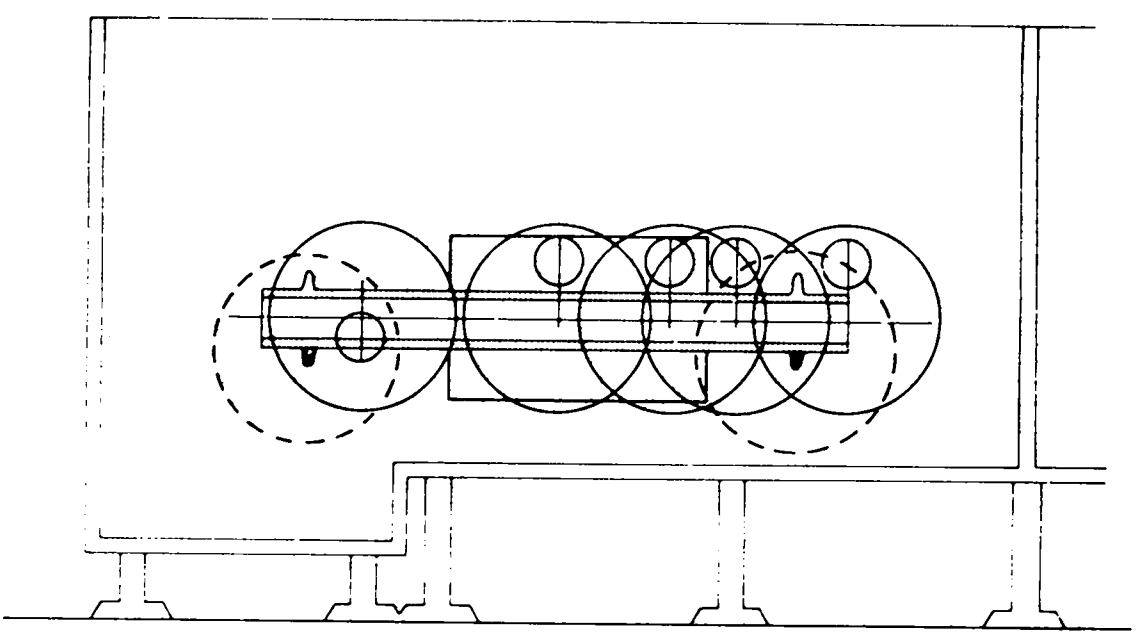

Fig. 7. Detector full fields of view for the five assay measurement locations (d = 63 cm) on the calciner axis (large solid circles), projected normal to the detector axis. Also shown are the fields of view (dashed circles) at the lower portion of each of the two bellows (shaded), projected normal to the detector axis. The corresponding assay (glove) port locations (small solid circles) are shown projected in the calciner vertical plane.

view) is approximately achieved for these measurement locations. The addition of a lower glove-port assay location (indicated in Fig. 4 with a "?" centered in a square grid shading of the glove port at the bottom of the figure left of center) is proposed to fill the gap in the "sampling" scheme.

Figure 8 is a plot of the radial dependence of the relative detection efficiency at the actual calciner measurement distance. The upper portion of the figure contains a sketch of a plutonium line source, fabricated of 1-in. Schedule 40 stainless-steel pipe and filled with 1342 g of plutonium in the

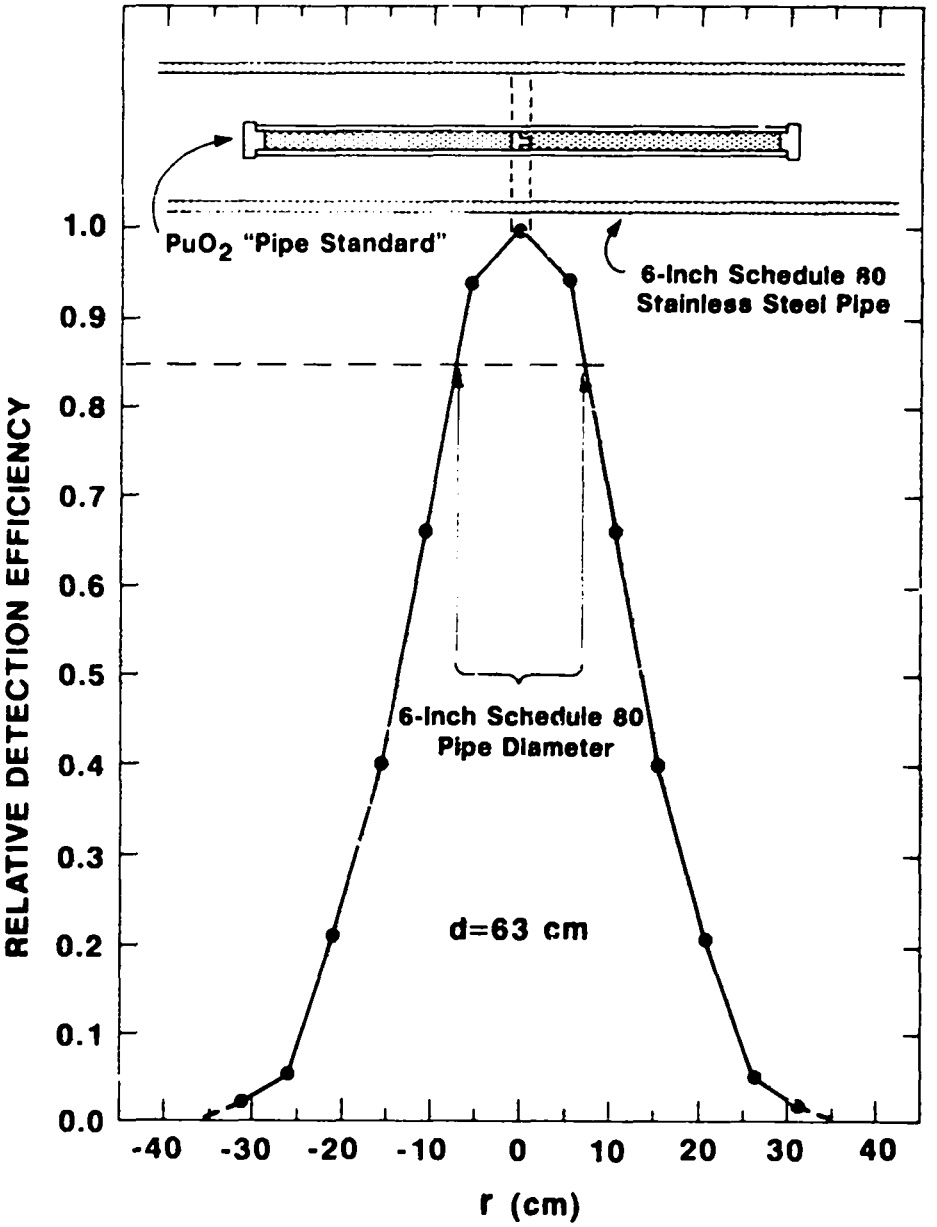

Fig. 8. Radial dependence of detection efficiency at d = 63 cm, the distance between the calciner center and the HPGe crystal for the assay geometry. The calciner tube and the PuO_2 line source are also drawn to the scale defined by the horizontal axis.

form of well-characterized PuO_2. This line source is drawn to the scale defined by the horizontal axis in Fig. 8. Its length (56 cm) effectively spans the detector field of view at the measurement distance. This source was built as a transmission source for measuring the attenuation effects of the hardware on the multiple-energy gamma-ray intensities. The equipment attenuation factors are determined by inserting the line source into the calciner tube, positioning it at the center of the calciner axis in the center of the detector field of view (for each measurement location), and measuring the equipment-attenuated intensities, I_{EQ}, at each gamma-ray energy. These intensities are compared with those determined by measurements in the same geometry with the detector viewing the line source removed from the calciner to give the I_0 values for each energy. The equipment attenuation correction factor for each gamma-ray energy is

$$CF_{EQ} = I_0/I_{EQ} \quad . \tag{1}$$

These correction factors are unique to each assay location. They are applied to the gamma-ray count rates measured at each assay location. The resulting assay values become independent of gamma-ray energy if there is no self-attenuation.

Figure 9 is a plot of one of the gamma-ray spectra acquired (viewing the calciner from the rightmost measurement location indicated in Fig. 4) in a 400-s count taken during the exercises. The spectrum energy ranges from 0 to 240 keV and 240 to 480 keV in Figs. 9(a) and 9(b), respectively. The assay was calibrated for the ^{239}Pu photopeaks at 129.3, 203.5 [Fig. 9(a)], 345.0, 413.7, and 451.5 keV [Fig. 9(b)], labeled in the figure. The quality of the spectra obtained at all locations on the calciner is equal to the spectrum quality obtained with the detector viewing the calciner at the feed end, a result in part of detector collimation, shielding and spectrum filtering. The ^{239}Pu photopeak at 375.0 keV (at channel 1890) appears prominently and free of (the 376.6-keV ^{241}Am) interference in all calciner spectra. The spectra obtained with plutonium standards, measured for calibration and verification purposes, display a significant 376.6-keV interference with the 375.0-keV photopeak. The significant presence of americium in the calciner feed material is

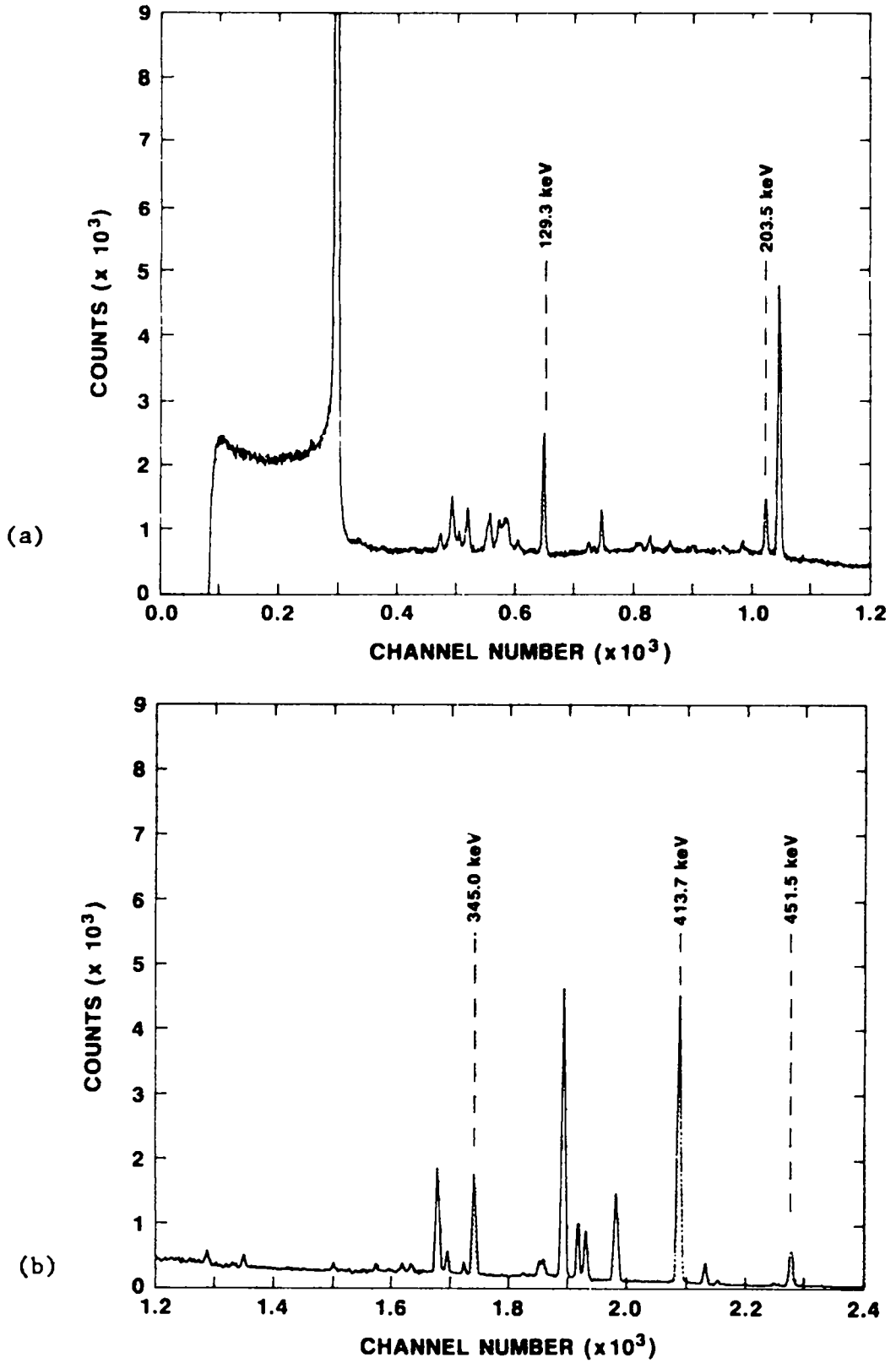

Fig. 9. Gamma-ray spectrum obtained at the calciner assay location at the feed end (rightmost measurement location in Fig. 2). The energy ranges of 0 to 240 keV and 240 to 480 keV are displayed in (a) and (b), with the ^{239}Pu assay peaks labeled.

not likely because of the nitrate ion exchange processing that immediately precedes precipitation and calcination. Therefore, the assay should include the 375.0-keV photopeak in future measurements.

IV. CALIBRATION AND VERIFICATION OF CALIBRATION

The point-source technique[2] was used at Los Alamos to calibrate the hold-up assay for the generalized (point, line, and area) deposit geometries. The point source, SPH-1, a small (5.348-g) plutonium metal sphere with 93.5% ^{239}Pu, was moved along a line (at 2-cm intervals) perpendicular to the detector axis and 24.4 cm from the HPGe crystal surface to obtain the calibration data. A 600-s count period was used for each measurement. The reference mass for the metal sphere was obtained by correcting (separately, for each gamma-ray assay energy) the ^{239}Pu mass for attenuation by the thin steel capsule walls and for self-attenuation by the plutonium metal. The correction factor for self-attenuation by a sphere is obtained from the fraction of gamma rays escaping from a sphere. This fraction takes the form[3]

$$F_{SPH} = (3/2Z)[1 - 2/Z^2 + e^{-Z}(2/Z + 2/Z^2)] \quad , \tag{2}$$

where

$$CF_{SPH} = (F_{SPH})^{-1} \tag{3}$$

is the "sphere" correction factor. The variable Z is the product of μ (the plutonium mass-attenuation coefficient), ρ (the plutonium density), and D (the diameter of the sphere). Table I gives the data used to obtain the reference values [energy-dependent effective ^{239}Pu masses, (M_e)] for the calibration.

The calibration was verified for point and line geometries at Los Alamos using various plutonium standards. One of the point standards was SPH-1. Another was a thin, aluminum-clad, plutonium metal foil containing 93% ^{239}Pu

TABLE I

COMPUTATION OF EFFECTIVE ^{239}Pu MASSES
FOR CALIBRATION OF MULTIPLE-ENERGY GAMMA-RAY HOLDUP ASSAY USING SPH-1

E (keV)	129.3	203.5	345.0	413.7	451.5
f_{239}	←		0.935		→
M_{Pu} (g)	←		5.35		→
μ (cm^2/g)	3.716	1.258	0.386	0.268	0.231
ρ (g/cm^3)	←		19.62		→
D (cm)	←		0.8045		→
$\mu \rho D = Z$	58.654	19.857	6.093	4.230	3.646
F_{SPH}	0.0256	0.0751	0.2331	0.3180	0.3570
μ_{Fe} (cm^2/g)	0.222	0.133	0.0983	0.0898	0.0862
ρ_{Fe} (g/cm^3)	←		7.860		→
x_{Fe} (cm)	←		0.0254		→
F_{Fe}[a]	0.9566	0.9738	0.9806	0.9822	0.9829
M_e (g ^{239}Pu)[b]	0.122	0.366	1.144	1.563	1.756

[a] $F_{Fe} = \exp(-\mu_{Fe} \rho_{Fe} x_{Fe})$.

[b] $M_e = M_{Pu} f_{239} F_{SPH} F_{Fe}$.

with 0.635 of plutonium (JTO 152801). The line standards were Zircaloy-clad mixed-oxide fuel rods (ROD 1063-23 and ROD 1063-7), each containing 29.93 g of plutonium (78.01% ^{239}Pu) in PuO_2 form, distributed uniformly over an active length of 121.7 cm, with a uranium-to-plutonium ratio of 42.4. The point and line calibrations were used to assay these standards to give the ^{239}Pu mass and mass-per-unit-length, respectively, at each assay energy. The SPH-1 point-source assay results were corrected for self-attenuation with the "sphere" correction factor, CF_{SPH}, Eq. (3). The JTO 152801 point-source assay results were corrected for self-attenuation with the "slab" correction factor,[4]

$$CF_{SLAB} = \mu\rho D/[1 - \exp(-\mu\rho D)] \quad , \tag{4}$$

where D is the slab (foil, in this case) thickness. The point-source assays were also corrected for attenuation by the capsule or cladding using the "absorber" correction factor,[4]

$$CF_{ABS} = \exp(\mu_{ABS}\, \rho_{ABS}\, D_{ABS}) \quad , \tag{5}$$

where μ_{ABS}, ρ_{ABS}, and D_{ABS} are the mass attenuation coefficients, densities, and thicknesses, respectively, of the cladding material. For iron- and aluminum-clad point sources, these correction factors are CF_{Fe} and CF_{Al}, respectively. The line-source assay results for the fuel rods were corrected for self-attenuation using the "cylinder" correction factor,[4]

$$CF_{CYL} = (\pi\mu\rho D/4)/[1 - \exp(-\pi\mu\rho D/4)] \quad , \tag{6}$$

where D is the cylinder (fuel rod pellet, in this case) diameter. The line-source assay results were also corrected for attenuation by the zirconium cladding using Eq. (5) to give CF_{Zr}. Table II summarizes the verification results obtained with the standards, showing reasonable agreement between the corrected

TABLE II

VERIFICATION OF HOLDUP ASSAY CALIBRATION WITH POINT AND LINE STANDARDS

	Eγ (keV)				
	129.3	203.5	345.0	413.7	451.5
SPH-1 (d = 19.3 cm)					
CF_{SPH}	39.12	13.31	4.29	3.14	2.80
CF_{Fe}	1.045	1.027	1.020	1.018	1.017
(g ^{239}Pu)					
A (1σ)	0.126 (0.006)	0.427 (0.037)	1.17 (0.06)	1.55 (0.04)	1.75 (0.17)
A_c (1σ)	5.07 (0.25)	5.64 (0.51)	5.12 (0.26)	4.95 (0.13)	4.98 (0.48)
R[a]	← 5.00 →				
A_c/R (1σ)	1.01 (0.05)	1.17 (0.10)	1.02 (0.05)	0.99 (0.03)	0.99 (0.10)
JTO 152801 (d = 24.6 cm)					
CF_{SLAB}	2.205	1.348	1.100	1.069	1.060
CF_{Al}	1.033	1.028	1.023	1.021	1.020
(g ^{239}Pu)					
A (1σ)	0.269 (0.002)	0.424 (0.008)	0.532 (0.011)	0.532 (0.007)	0.474 (0.026)
A_c (1σ)	0.613 (0.005)	0.588 (0.011)	0.598 (0.012)	0.581 (0.008)	0.512 (0.029)
R[b]	← 0.591 →				
A_c/R (1σ)	1.04 (0.01)	0.99 (0.02)	1.01 (0.02)	0.98 (0.01)	0.87 (0.05)
ROD 1063-21 and ROD 1063-7 (d = 24.6 cm)					
CF_{CYL}	31.0	10.4	3.42	2.59	2.33
CF_{Zr}	1.445	1.135	1.066	1.054	1.050
(g ^{239}Pu/cm)					
A (1σ)	0.0089 (0.0003)	0.0353 (0.0022)	0.133 (0.003)	0.132 (0.001)	0.134 (0.009)
A_c (1σ)	0.399 (0.013)	0.417 (0.026)	0.485 (0.011)	0.360 (0.003)	0.328 (0.022)
R[c]	← 0.384 →				
A_c/R (1σ)	1.04 (0.03)	1.08 (0.07)	1.26 (0.03)	0.94 (0.01)	0.85 (0.06)
Rocky Flats Pipe Standard (d = 60 cm)					
CF_{CYL}	36.1	12.3	3.95	2.91	2.61
CF_{SLAB}	46.0	15.7	4.96	3.57	3.17
CF_{St}	1.79	1.42	1.29	1.26	1.25
(g ^{239}Pu/cm)					
A_0 (1σ)	0.12 (0.02)	0.9 (0.1)	3.3 (0.1)	4.6 (0.1)	5.2 (0.3)
$A_{c,CYL}$ (1σ)	8 (1)	15 (2)	17 (1)	17 (0)	17 (1)
$A_{c,SLAB}$ (1σ)	10 (2)	20 (2)	21 (1)	21 (0)	21 (1)
R[d]	← 20 →				
$A_{c,CYL}/R$ (1σ)	0.4 (0.1)	0.8 (0.1)	0.9 (0.1)	0.9 (0)	0.9 (0.1)
$A_{c,SLAB}/R$ (1σ)	0.5 (0.1)	1.0 (0.1)	1.1 (0.1)	1.1 (0)	1.1 (0.1)

[a] R = 5.35 (g Pu) • 0.935 (g ^{239}Pu/g Pu) = 5.00 g ^{239}Pu.

[b] R = 0.635 (g Pu) • 0.930 (g ^{239}Pu/g Pu) = 0.591 g ^{239}Pu.

[c] R = 2 • 29.93 (g Pu) • 0.7801 (g ^{239}Pu/g Pu) ÷ 121.7 cm = 0.384 g ^{239}Pu/cm.

[d] R = 0.9 • 1342 (g Pu) • 0.93 (g ^{239}Pu/g Pu) ÷ 56 cm = 20 g ^{239}Pu/cm.

assay results and the reference values for the standards. The distance between the HPGe crystal and the source is d, the reference values are R, the point- and line-source assay results are A, and the corrected assay results are A_C. The average (over the five assay energies) ratio of the corrected assay result to the reference value, A_C/R, is 1.04 (1σ = 0.08) for SPH-1, 0.98 (1σ = 0.06) for JTO 152801 and 1.03 (1σ = 0.15) for ROD 1063-23 and -7. Thus, the calibration is verified to better than 10% across the range of assay energies. The agreement improves at 413.7 keV, where the precision for the calibration measurements and the verification measurements is the best.

At Rocky Flats, the PuO_2 line source inside the glove box was assayed bare to verify the line-source holdup calibration before actual measurements were performed. The assay distance was 50 cm, and the source was horizontal. The bare source assay results, A_0, are given for each gamma-ray assay energy in Table II. Because the PuO_2 powder that was used to make the source filled the pipe to only 85% of its capacity, the detector viewed a powder geometry between that of a cylinder and a slab. Therefore, there is some uncertainty about the form of the correction factor for self-attenuation. Furthermore, because the standard was fabricated in two pieces, a 3-cm-wide "dead" spot (containing no PuO_2) exists at the very center where the two pieces join. At an assay distance of 60 cm, this gap should reduce the effective reference value by $\lesssim$10%. (Refer to Fig. 8.) The holdup assay results were corrected for self-attenuation using both "cylinder" and "slab" correction factors computed for the PuO_2 contents from Eqs. (6) and (4), respectively, and for attenuation by the steel pipe computed from Eq. (5) to give CF_{St}. Table II summarizes the verification results obtained with the pipe standard showing that the corrected assay results obtained with cylinder correction factors and those obtained with slab correction factors bracket the reference value for the PuO_2 pipe standard, as expected, except for the result at 129 keV for which the attenuation effect of the lead-impregnated glove (not included in the corrections) is the probable source of the apparent negative bias in the corrected assays.

V. CALCINER MEASUREMENTS

Line-source holdup assays were performed at each of the five calciner measurement locations as indicated by the detector locations shown in Fig. 4

and the corresponding calciner fields of view shown in Fig. 7. Multiple (4 ± 1) assays of 400 s each, with repositioning of the detector and significant elapsed time between assays, were performed at each assay location to establish uncertainties associated with such variables as positioning and changing background. The average assay results, $\bar{A}$, and the standard deviation in the multiple assays, σ_A, are given (for each energy) for each location in Table III, where the measurement locations 1 through 5 correspond to the consecutive locations from left to right indicated in Figs. 4 and 7. The $\bar{A}$ values for a given location are energy-dependent: the expected result of attenuation effects.

With the detector positioned as for the calciner assays, the PuO_2 line-source was inserted into the calciner, and centered at the detector field of view for each measurement location. The difference between the resulting line-source assay result (obtained in a 400-s count) and the corresponding result, A, obtained without the line source in place, was divided into the bare source assay result, A_0, for each gamma-ray energy (as given in Table II) giving, according to Eq. (1), the energy-dependent equipment attenuation correction factors at each measurement location. These are the CF_{EQ} values in Table III.

The strongly energy-dependent equipment attenuation correction factors are large and variable, ranging from 4 to 20 at 414-keV . The uncertainties in the CF_{EQ} values, especially at the other assay energies and in particular at location 1, are very large. This is the result of the statistical effects of obtaining the small difference of two large numbers. The effect is greatly enhanced as equipment attenuation increases (at locations 3 and 4), as gamma-ray energy decreases, and as holdup deposits dominate the count rate (at location 1) increasing the background level to equal or exceed the magnitude of the PuO_2 line-source contribution to the signal. At location 1, the line-source contribution could not be distinguished above the background. Hence, the CF_{EQ} value was determined as a lower limit obtained from the 3σ value in the difference between the A values measured with and without the line source, where the 3σ value is defined as the upper limit of the line-source contribution to the signal. Visible loose accumulations within the calciner at the feed end were a major contributor to the high-background level in the assays at this location.

TABLE III

CALCINER LINE SOURCE ASSAY RESULTS IN g ^{239}Pu/cm

Eγ (keV)	129.3	203.5	345.0	413.7	451.5
Location 1 (feed end)					
$\bar{A}$ (σ_A)	0.36 (0.02)	2.07 (0.21)	5.41 (0.17)	6.72 (0.09)	7.09 (0.36)
CF_{EQ} (σ_{CF})	≥1	≥1	≥2.2	≥5.8	≥1.6
A_c[a] (σ)	≥0.72 (0.04)	≥4.1 (0.4)	≥23.8 (0.8)	≥78.0 (1.0)	≥22.7 (1.2)
Location 2					
$\bar{A}$ (σ_A)	0.06 (0.02)	0.21 (0.05)	0.52 (0.10)	0.61 (0.14)	0.69 (0.26)
CF_{EQ} (σ_{CF})	12 (18)	6.0 (3.0)	7.4 (0.2)	8.1 (0.6)	14.2 (9.2)
A_c (σ)	0.7 (1.1)	1.3 (0.7)	3.9 (0.7)	5.0 (1.2)	9.8 (7.3)
Location 3					
$\bar{A}$ (σ_A)	-0.01 (0.01)	0.17 (0.05)	0.55 (0.18)	0.69 (0.26)	0.67 (0.43)
CF_{EQ} (σ_{CF})	≥3	≥5.3	29 (22)	20 (4)	16 (8)
A_c (σ)	≥-0.03 (0.03)	≥0.9 (0.3)	16 (13)	14 (6)	11 (9)
Location 4					
$\bar{A}$ (σ_A)	0.04 (0.02)	0.05 (0.04)	0.17 (0.09)	0.16 (0.07)	0.4 (0.3)
CF_{EQ} (σ_{CF})	≥3	90 (450)	16 (5)	12 (1)	24 (22)
A_c (σ)	≥0.1 (0.1)	5 (23)	2.7 (1.3)	2.0 (0.9)	9 (11)
Location 5 (product end)					
$\bar{A}$ (σ_A)	0.13 (0.01)	0.31 (0.04)	0.39 (0.05)	0.45 (0.08)	0.35 (0.13)
CF_{EQ} (σ_{CF})	12 (13)	6 (3)	4.1 (0.5)	3.78 (0.14)	3.3 (0.5)
A_c (σ)	1.6 (1.7)	1.7 (0.8)	1.6 (0.3)	1.7 (0.3)	1.2 (0.5)

[a]At location 1 only, $A_c = 2 \cdot A \cdot CF_{EQ}$. (At other locations, $A_c = A \cdot CF_{EQ}$.)

The magnitudes of the CF_{EQ} values at 414 keV (where these results are most precise) can be compared to calculated minimum correction factors obtained from the known uniform components of the calciner structure. These components are the 6-in. (15-cm) schedule 80 steel pipe, the inner tube of the calciner that extends over its entire length, and the 15.2-cm-thick fire brick heat shield that surrounds the 90-cm-long heater section of the calciner. These components define the minimum attenuation because the schedule 80 tube is overlaid with sleeves of varying lengths and thicknesses and fitted with numerous flanges and mounting plates all along its length. The heat shield too is encased in steel on its inner and outer surfaces and is fitted with other hardware. The absorber correction factors, computed from Eq. (5) for 414-keV gamma rays from the known compositions and thicknesses of the calciner tube and heat shield, are 2.3 and 4.3, respectively. Deposits in the "heater" section of the calciner are attenuated at a minimum by a product of these two correction factors (2.3 x 4.3 = 10.0) and those in the "tube" section are attenuated by a minimum factor of 2.3. Table IV compares these minimum expected equipment attenuations, $CF_{EQ,MIN}$, at each location (where Fig. 5 is used to determine whether the "heater" or "tube" values apply) with the measured results. The measured attenuations are approximately twice the minimum values, an entirely reasonable result for this calciner structure.

TABLE IV

COMPARISON OF MEASURED EQUIPMENT ATTENUATION CORRECTIONS WITH MINIMUM EXPECTED VALUES FOR 414-keV GAMMA RAYS

Calciner Assay Location	Heater (H) or Tube (T) Section	CF_{EQ}	$CF_{EQ,MIN}$	$\frac{CF_{EQ}}{CF_{EQ,MIN}}$
1	T	$\geq$5.8	2.3	$\geq$2.5
2	T (and H)	8.1 ± 0.6	>2.3	<3.5 ± 0.3
3	H	20 ± 4	10.0	2.0 ± 0.4
4	H (and T)	12 ± 1	<10.0	>1.2 ± 1.0
5	T	3.8 ± 0.2	2.3	1.6 ± 0.1

It is clear that improved values of CF_{EQ} are accessible if the transmission measurements could be performed without time constraints and on clean equipment (after a disassembly). Because these values need not be remeasured once established, and because each assay depends on these values, a significant expenditure of time in obtaining these values is justifiable.

Finally, although the 414-keV value of CF_{EQ} at location 1 is given as an upper limit, it is derived from the most precise data obtained at this location. Therefore, it is not too surprising that the ratio of this limit to the minimum estimate is similar in magnitude to the ratios at the other locations, as seen in Table IV.

VI. EVALUATION OF CALCINER HOLDUP

The average line-source assay results, $\bar{A}$, are multiplied by the equipment attenuation correction factors, CF_{EQ}, to give the corrected assay results, A_C, in g ^{239}Pu/cm in Table III. Because the calciner covers only half of the detector field of view at location 1 (refer to Fig. 7), the A_C results for location 1 (only) have been multiplied by 2.

The unreasonably large uncertainty in the CF_{EQ} values, particularly as the gamma-ray energy decreases, destroys the sensitivity of the energy diagnostic as an indicator of (uncorrected) self-attenuation effects. Within the large uncertainties in these values, the A_C results are independent of energy at each location, but more precise correction factors must be obtained for a meaningful evaluation. It is clear from having directly physically identified substantial loose accumulations at the feed end of the calciner tube, that residual (self-) attenuation effects impose a negative bias on the assay results for location 1. This would be obvious (from the energy dependence of the A_C results at location 1) if precise CF_{EQ} values were available.

The low-energy assay results suffer a two-fold disadvantage. The CF_{EQ} values are less precise and the $\bar{A}$ values are less accurate, primarily because the equipment attenuation is so large that background uncertainties (including the effects of surface contamination) can be significant. From this point of view, the ability to use the 375.0-keV gamma ray of ^{239}Pu in the holdup assay becomes more appealing.

The validity of the line-source assumption in the holdup assay was examined at the two locations (feed and product ends of the calciner, locations 1 and 5, respectively) where concentrated accumulations are expected. Figure 7 shows two darkened spots, corresponding to the lower portions of the Teflon bellows, where material becomes lodged and inaccessible to cleanout except during disassembly. Recent modifications in the calciner design have been aimed at reducing accumulations in the bellows. However, some efforts were made to determine the possible existence of a (dominant) point source at each of the two locations. Separate measurements were performed at the two measurement locations (1 and 5) with the detector field of view centered on the bellows bottom (rather than on the calciner axis) to enhance the possible point-source effect. The corresponding fields of view appear as large dashed circles in Fig. 7. Measurements were performed at two assay distances (65 cm and 50 cm) to evaluate the d-dependence of the signal. The dependence fell between d^{-1} and d^{-2} at both locations indicating effective deposit geometries somewhere between line- and point-source distributions. At the product end, the point source (in the bellows) is likely to be small enough that accumulations in the tube compete significantly, causing the ambiguity in the deposit geometry. At the feed end, bellows accumulations are expected to be larger. However, the known, large, visible deposit at the bottom of the calciner tube in the extreme 15-cm portion of the feed end is the probable cause of the masking of a d^{-2} dependence that might result from a large deposit in the bellows. For the future, the cleanout of loose accumulations and gross deposits from the calciner tube (short of a disassembly) should be complete before the holdup measurements are performed. The bellows bottoms will be shielded from the detector during the on-axis (line-source) assays of the calciner holdup. Finally, the bellows component will be measured separately as a point source in a more closely coupled geometry, and a 10-mCi ^{137}Cs transmission source will be used to evaluate the self-attenuation correction for the bellows accumulation.

Assuming that the line-source approximation is reasonable for the measured calciner deposits, the corrected assay results, A_c, (from Table III) can be used to determine the calciner holdup. The procedure is to average the assay results for the (five) measurement locations and apply the average to the total calciner length. Because the calciner tube is included in only one-half of the detector field of view for the measurement at location 1, this

assay result has been given a 50% weighting relative to each of the other four. The calciner holdup assay is

$$H(g\ Pu) = \bar{A}_c \bullet L \div 0.93 \quad , \tag{7}$$

where $\bar{A}_c$ is the (weighted) average corrected line-source assay result (in g ^{239}Pu/cm), L is 224 cm, the length of the calciner tube, and 0.93 is the ^{239}Pu fraction. The weighted average corrected line-source assay, computed from 414-keV $A_c(i)$ results at the five locations, i, is

$$\bar{A}_c = [\tfrac{1}{2} A_c(1) + \sum_{n=2}^{5} A_c(n)] \div 4.5 \tag{8}$$

$$\geqq 13.7 \pm 1.4\ g\ ^{239}Pu/cm \quad .$$

Substitution of this result into Eq. (7) gives

$$H \geqq 3300 \pm 340\ g\ Pu \quad .$$

The feed end (location 1) component of this result is

$$H_{FEED} \geqq 2090 \pm 30\ g\ Pu \quad ,$$

with the remainder,

$$H_{REM} = 1210 \pm 340$$

ascribed to the portion of the calciner downstream of the feed measurement location.

The H_{FEED} value is a lower limit for two reasons. The first is that the CF_{EQ} measurement was estimated at its minimum value. A remeasured result for CF_{EQ} at location 1 after cleanout can be applied subsequently to the $\bar{A}$ values to give an absolute measurement with an uncertainty for H_{FEED}. However, it is believed that this measurement will be biased low because of the self-attenuation of the 414-keV gamma rays in the substantial loose accumulations at the feed end of the calciner.

A calciner cleanout was performed following these measurements. The recovered amounts of 4175 g of plutonium at the feed end and 1375 g of plutonium at the discharge end are consistent with the measured results. An additional 970 g of plutonium was recovered from the glove-box floor. It is not clear whether this was a component of the assay (originating within the calciner tube) or whether this material was on the calciner floor at the time that the holdup measurements were performed.

VII. CONCLUSIONS AND RECOMMENDATIONS

The first field testing of high-resolution gamma-ray methods and current hardware and software technologies applied to generalized geometry measurements of solid plutonium holdup in high-throughput bulk-processing equipment has provided promising results. Because of automation, the calibration and the holdup measurements are reasonable for the multiple-energy approach. New equipment and detector designs have reduced sizes and weights to meet practical needs in these applications. The measurement results, while imprecise, are in reasonable agreement with cleanout values. Furthermore, there is a good prospect that the measurement precision will improve dramatically if the correction factors for equipment attenuation can be remeasured (with ample

time allotted for the measurements) after reassembly of the calciner following the cleanout.

Certain criteria must be satisfied before the holdup measurements are performed to maximize the chances of the success of these measurements for the Rocky Flats calciner. Most of these criteria (those marked with an asterisk, *, below) were satisfied for the recent holdup measurements, indicating a significant commitment on the part of the facility operator to achieving success in these efforts. While the criteria are worded to address the Rocky Flats calciner specifically, they can be generalized to apply to most gamma-ray holdup measurements of high-throughput bulk-processing equipment. The criteria are:

(1) Plutonium-bearing material should be removed from the process equipment cavities to the extent possible, short of a complete disassembly.

*(2) Auger feed assembly and the product end plate must be removed to permit (visual and physical) access to the inner tube from either end.

(3) Tumbler rods must be removed entirely from the inner tube.

*(4) Glove-box product-end viewing window must be in place.

*(5) Contaminated equipment (such as the auger feed mechanism or the tumbler rods) should be cleaned or removed to a remote location. An adjacent glove box is acceptable.

(6) Equipment and glove-box surfaces including floors should be wiped down (as for inventory), and the cleanup scrap removed from the glove box.

*(7) <u>It is desirable</u> to have relatively new gloves on the (seven or eight) glove ports used for the measurements.

*(8) Bulk-storage locations adjacent to the calciner line should be empty.

*(9) The 56-cm-long PuO_2 standard should be available in the line. (Eventually, this need will be phased out.)

(10) A device for positioning the PuO_2 standard within the inner tube must be available. The device must

(a) locate the standard at the radial center of the inner tube.

(b) allow for a known positioning of the standard at any location along the length of the tube.

(c) be usable (with the standard in place) while the tube is rotating, <u>if possible</u>.

(Note that this positioning device will eventually be phased out when the need for the standard is phased out. Refer to 9.)

*(11) A knowledgable operator must be available to assist with measurements.

*(12) A clean area must be available for safe measurement equipment storage.

(13) A 10-mCi ^{137}Cs point source must be available for transmission measurements.

The plan is set to remeasure the equipment attenuation correction factors for the calciner after reassembly following the cleanout. The measurement of these attenuation correction factors for the Rocky Flats hydrofluorinator, nearly identical in design to that of the calciner, will also be scheduled if time permits, following cleanout and reassembly of the hydrofluorinator. The

criteria listed above for calciner measurements apply directly to the Rocky Flats hydrofluorinator.

Because of production delays associated with the scheduling of measurements on the cleaned and reassembled equipment, the measurements of equipment attenuation effects performed on active processing equipment will necessarily be constrained by time. Future holdup measurement needs must be anticipated before production equipment is commissioned so that such measurements can be performed free of serious access and time limitations.

The generalized-geometry assay approach has shown promise in this difficult holdup measurement application. The automation of this approach greatly enhances the potential facility-wide usefulness. Therefore, the prospects are good for extending the generalized geometry assay approach to other holdup measurement needs.

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