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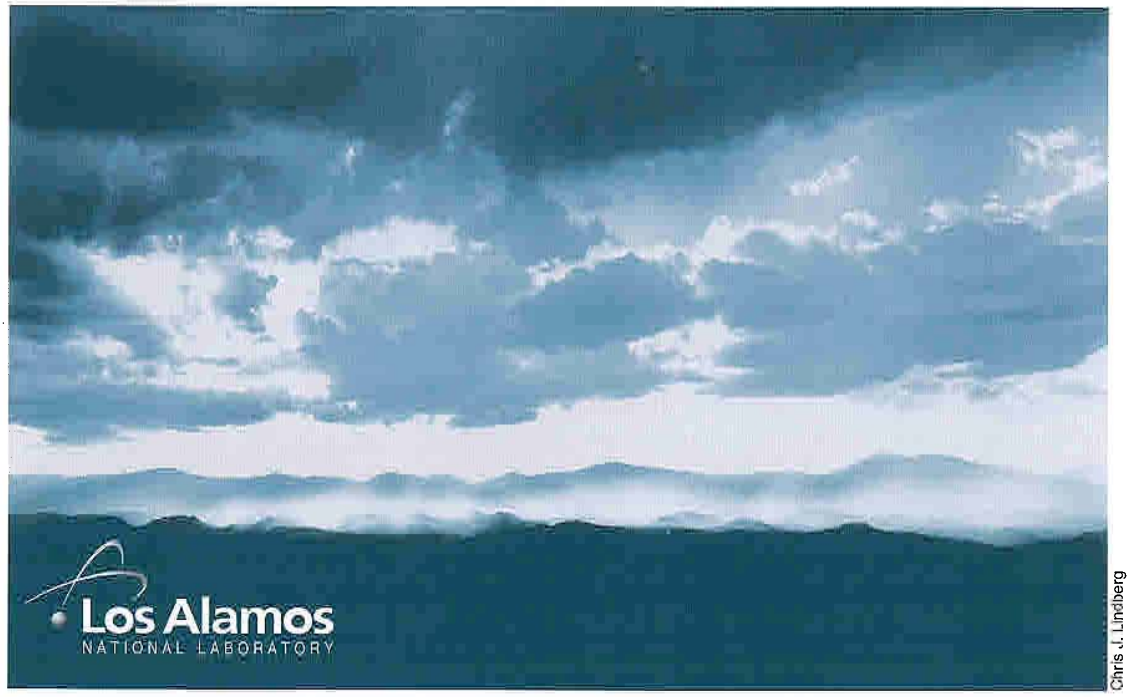

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ABSTRACT

A recent effort determined uranium holdup at a large fuel fabrication facility abroad where low enriched (~3%) uranium (LEU) oxide feeds the pellet manufacturing process. Measurements taken with both high-and low-resolution gamma-ray spectrometry systems include extensive data for the ventilation and vacuum systems. Equipment dimensions and the corresponding holdup deposit masses are large for LEU. Because deposits are infinitely thick to the 186 keV gamma ray in many locations in an LEU environment, measurements of both the 186 and 1001 keV gamma-rays were required, and self-attenuation was significant at 1001 keV in many cases. These wide-dynamic-range measurements used short count times, portable scintillator detectors, and portable MCAs. Because equipment is elevated above floor levels, most measurements were made with detectors mounted on extended telescoping poles. One of the main goals of this effort was to demonstrate and validate methods for measurement and quantitative analysis of LEU holdup using low-resolution detectors and the Generalized Geometry Holdup (GGH) techniques. The current GGH approach is applied elsewhere for holdup measurements of plutonium and high-enriched uranium. The recent experience is directly applicable to holdup measurements at LEU facilities such as the Paducah and Portsmouth gaseous diffusion enrichment plants and elsewhere, including LEU sites where D&D is active. This report discusses the measurement methodology, calibration of the measurement equipment, measurement control, analysis of the data, and the global and local assay results including random and systematic uncertainties. It includes field-validation exercises (multiple calibrated systems that perform measurements on the same extended equipment) as well as quantitative validation results obtained on reference materials assembled to emulate the deposits in an extended vacuum line that was also measured by these techniques. The paper examines the differences in assay results between the low-resolution system using the GGH method and the high-resolution system utilizing the commercially available ISOCS analysis method. This project is funded by the United States Department of Energy Office of International Safeguards (NA-243).

INTRODUCTION

Extensive measurements of low-enriched uranium (LEU) holdup in vacuum and ventilation lines were made at a foreign fuel-fabrication plant. The measurements were made using the Generalized Geometry Holdup (GGH) assay technique. The feed oxide for the pellet manufacturing process varies in enrichment from 1.0% to 4.45%. Currently this facility makes fuel pellets for the RBMK and VVER reactors. The plant also produces ceramic grade UO_2 powder, which it sells.

OVERVIEW OF THE GGH METHOD AND MODELS

The GGH method is described in detail elsewhere [1]. The following is a brief overview of the technique and its models. Holdup measurements are usually made with collimated, shielded gamma-ray detectors. The distance between the deposit and detector is chosen to approximate the geometry of the deposit to that of an ideal point, line, or area in a radially symmetric field of view. The quantitative result is based on the net count rate of a representative gamma ray from the isotope

of interest. Several corrections are applied to the measured rates, including those for the effects of room background and equipment attenuation effects. The GGH technique employs relatively new generalized procedures that correct for the effects of both finite-source dimensions and gamma-ray self-attenuation. The finite-source effect arises from the fact that point and line deposits are not ideal in that actual deposits have finite (non-zero) width. Ignoring the finite width results in a negative bias in the isotope mass determined by a calibration that assumes an ideal deposit geometry. The self-attenuation correction is required because uranium is highly attenuating to its own gamma radiation. The self-attenuation effect increases non-linearly with deposit thickness and also results in a negative bias if ignored. Because of the non-linear dependence on the pre-correction deposit thickness, the self-attenuation correction must be applied last. One parameter provided by the user, the "Finite-source Dimension", applies to both corrections. This is the user's best guess of the width of a point or line deposit. After all corrections are made to the data a specific isotope mass is obtained in units of grams (g) for a point, g/cm for a line, or g/cm^2 for an area deposit. The total isotope mass of a line or area deposit is the specific mass multiplied by the deposit length or area, respectively.

MEASURING LEU HOLDUP

The 186-keV gamma ray is normally utilized for uranium holdup measurements because it is a direct measure of ^{235}U and has a moderate yield (~43,000 γ/s/g ^{235}U). It is often necessary to use the 1001-keV gamma ray to quantify uranium in LEU holdup. Three issues arise with this gamma ray:

1) Its yield is very low (~75 γ/s/g^{238}U).
2) The photoelectric absorption probability in a 5-cm-thick sodium iodide (NaI) crystal is only 1.3% compared to 91% for the 186-keV gamma ray. This also results in a significant Compton continuum at lower energies.
3) The 1001-keV-gamma ray is highly penetrating, rendering the detector shield much less effective and resulting in the need to measure and subtract a substantial room background at each measurement location.

Nevertheless, much thicker deposits can be measured with the 1001-keV gamma ray than with the 186-keV gamma ray. A deposit must be less than infinitely thick to the gamma ray in order to quantify the deposit in a strictly a passive measurement . Table 1 shows limiting thicknesses for the 186 and 1001-keV gamma rays for different uranium materials quantified using the passive gamma ray measurement.

Table 1. Limiting thicknesses (in grey) for various uranium materials.

		186-keV gammas		1001-keV gammas	
material	density: ρ	μ (186)	1/(μρ)	μ (1001)	1/(μρ)
	g/cm^3	cm^2/g	cm	cm^2/g	cm
metal	18.7	1.47	0.04	0.075	0.71
UF_6 (solid)	4.7	1.03	0.21	0.070	3.04
UO_2 (sintered)	10.9	1.31	0.07	0.074	1.24
UO_2 (powder)	2.0	1.31	0.38	0.074	6.76
U_3O_8 (highly packed)	7.3	1.26	0.11	0.073	1.88
$UO_2(NO_3)_2 \cdot H_2O$	2.8	0.76	0.47	0.071	5.03

These thicknesses were derived by taking into account the self-attenuation correction for a purely passive measurement. This correction takes the form of equation (1):

$$CF_{ATTEN} = const * \ln[1-\mu\rho x] \qquad (1)$$

where x is the thickness of the deposit. Thus, x cannot be greater than $1/(\mu\rho)$ in order to evaluate the natural logarithm in the correction. Deposits measured ranged from thicknesses thin enough to use 186-keV gammas to those for which even 1001-keV gammas were severely self-attenuated. While the 186-keV gamma ray has the advantage of better counting statistics, a combination of large attenuation effects and the presence of discrete interferences at the lower gamma-ray energy, as discussed below, enforces use of 1001-keV for quantitative analysis in many if not most situations in which LEU is measured.

EQUIPMENT USED

The following equipment was used for measurements of LEU holdup in July 2004:

- shielded, compact NaI detectors (2.5-cm diameter by 5-cm-thick crystals)
- Canberra InSpector 2000 MCAs w/ Genie 2000 software
- Laptop computers with GGH specific analysis spreadsheets
- poles and hardware to extend detectors to equipment for measurements
- tungsten plugs

The equipment used for measurements of LEU holdup performed in October 2004 was basically the same with the exception of the MCAs (GBS Elektronik MCA166) and acquisition software (WinSPEC). Figure 1 is a spectrum of a relatively thick deposit acquired during the campaign in October showing the regions of interest (ROIs) as red dashed lines. The tungsten plug is inserted into the detector collimator to measure the room background contribution at each measurement location. Therefore, two spectra were acquired at each measurement location, one without and one with the tungsten plug. The relatively large room background contribution at 1001-keV in Figure 1 emphasizes the difficulty of measurements performed at 1001-keV and the importance of performing the measurements of room background.

The 186 and 1001-keV gamma ray peaks are clearly visible in Figure 1. Also of great interest is the small peak at 238-keV from the decay of ^{212}Pb in the ^{232}U decay chain. The presence of ^{232}U indicates that this is recycled uranium. Because the 238-keV peak in NaI spectra is barely resolved from the 186-keV peak, ROIs for the peak and continuum must be set conservatively, as illustrated, to assure that 238-keV activity does not contribute to the continuum ROI above the 186-keV peak. The ROI boundaries for the 186-keV peak and continuum were: 159 – 199-keV and 201 – 210-keV respectively. The 1001-keV peak and continuum ROIs were: 943 – 1069-keV and 1075 – 1201-keV respectively. Liberal ROI limits are possible at 1001-keV because of the absence of interferences. Such settings are much more forgiving of gain drift.

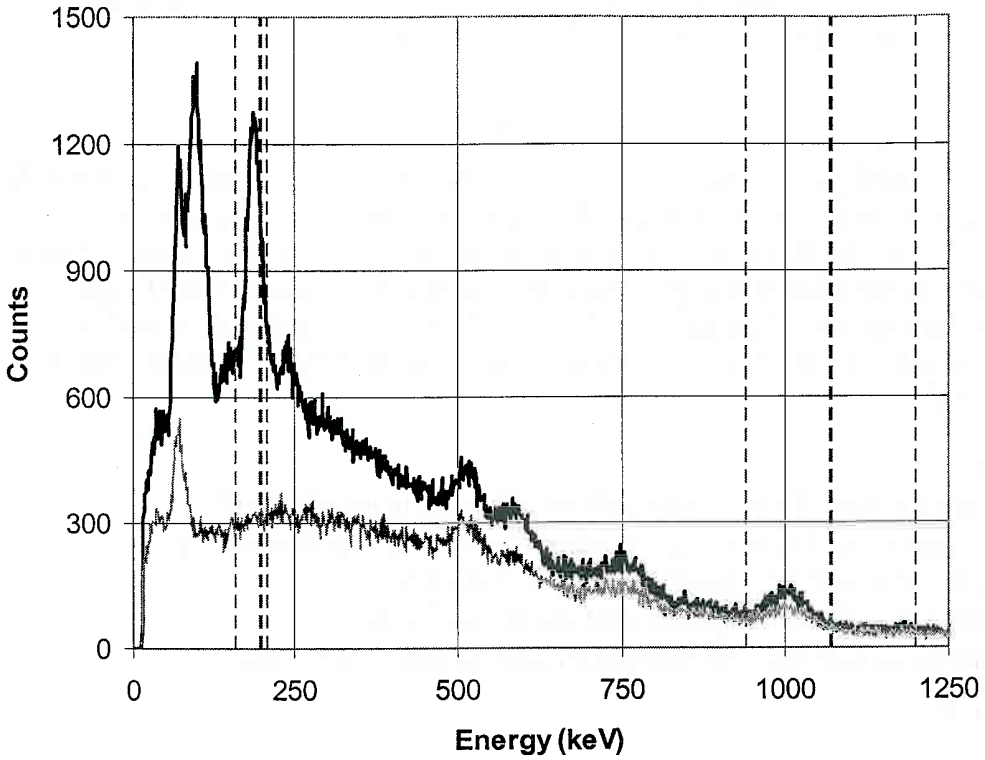

Figure 1. LEU spectrum showing the 238-keV peak interference. The red dashed lines show the boundaries of the regions of interest (ROIs). The grey spectrum is room background measured for the same count time with the tungsten plug inserted in the collimator.

EQUIPMENT CALIBRATION

LEU holdup deposits are often (while HEU holdup deposits are seldom) infinitely thick to the 186-keV gamma ray, therefore, data for 186- and 1001-keV gamma rays must be acquired simultaneously. Likewise, the detectors must be calibrated at both energies. Two types of measurements at each energy were performed at LANL before each trip to calibrate the holdup measurements. The first uses a point source reference standard at a well defined distance and at the center of the detector's field of view (on-axis). This is an absolute calibration that assumes a $1/r^2$ dependence of the detector response to a point source. The second type of measurement establishes the relative response of the detector to off-axis locations of the deposit within the field of view. This is necessary to apply the absolute calibration to extended (line and area) deposit geometries. The relative response is constructed by measuring a point source at discrete locations off the axis of the detector, and since it is a relative (-to-the-on-axis) measurement, it is not necessary that the source be a standard. It is necessary that the energy of the gamma ray be the same, or nearly so, as that measured for the absolute calibration. The relative off-axis responses were measured using the 186-keV gamma ray from a ^{235}U source and the 1064-keV gamma ray from a ^{207}Bi source.

HOLDUP DATA ANALYSIS

A spreadsheet was utilized with an embedded Visual Basic (VB) program to analyze the measurement data. There are two worksheets in this spreadsheet, one for 186-keV (^{235}U) data, and the other for 1001-keV (^{238}U) data. In addition to the measurement specific data (ROI integrals, count-time, measurement distance, etc.) the spreadsheet requires the weight fraction of the isotope. Once all required information is entered in the spreadsheet a button is clicked that executes the VB program. This program takes the room-background-subtracted net count rate for the gamma-ray of interest, corrects for container wall attenuation, and then calculates a specific mass for that isotope using the appropriate calibration constant and distance multiplier, if any. This specific mass is then first corrected for finite-source effects and second for self-attenuation[1]. The program then calculates the total uranium holdup mass by dividing the corrected specific mass by the weight fraction of the isotope and multiplying by length of the extended line or by the area of the extended surface as appropriate. The program propagates the random uncertainty from counting statistics through the analysis algorithms to give the random uncertainty (σ) in the holdup mass. The program also performs tests on the data to determine if the deposit is thicker than or within 3σ of the limiting thickness. If the deposit is greater than the limiting thickness then the self-attenuation correction cannot be performed, and no final mass is calculated for that measurement. If the deposit is thin, but is within 3σ of the limiting thickness the user should be cautious of the final results because the self-attenuation correction for these deposits is likely to be large and steeply changing with thickness, causing a sharp increase in the relative measurement uncertainty propagated through the correction algorithm.

Typically for extended line and area deposits, several measurements are taken at different locations. A specific mass is calculated for each measurement that is entered in the spreadsheet. The spreadsheet can average the specific masses for the extended line or area deposit and assign that average to the entire deposit. By averaging several measurements of the same extended deposit the final relative uncertainty in the average is smaller than those of the individual measurements. Short counts allow measurements at more deposit locations, providing better sampling of the extended deposits. The statistics for one long count are equivalent to those for the average of many short counts that sum to the same count time.

VERIFICATION AND MEASUREMENT CONTROL

Each detector's absolute calibration was verified daily using HEU and depleted uranium (DU) sources of known uranium mass and isotopic composition provided by the facility. The radial response of the detectors was not verified at the plant due to time considerations. The calibration verifications were all within 14% except for one detector whose calibration at 186-keV was low by 45%. This was later shown to be the result of a gain drift.

Each morning the 186-keV peak count rate, centroid (gain), and full width at half maximum (FWHM) were measured using the Ulba HEU source to ensure a consistent response from day to day. Each team also routinely verified the gain and FWHM after every 12th measurement in the plant. Some drift was observed, and appropriate gain corrections were made.

EXAMPLE OF MEASUREMENT RESULTS

The contents of one of the measured vacuum lines are static. This line has been measured several times with the GGH method utilizing low-resolution detectors (NaI) and the 1001-keV gamma ray. It has also been measured multiple times[2] using ISOCS[3], a method that employs a high-

resolution Ge detector. The line itself is 74 m long with a diameter of 9 cm. The measurement distance of 40 cm was suitable for GGH analysis of line deposits in this case. The distance between measurement points was chosen to be 1 m. The line was measured by three groups in July 2004 and by two groups in October 2004 using the GGH method. It was also measured three times using the ISOCS methodology. The total uranium mass result and 1σ random uncertainty for each measurement are given in Table 2. It should be noted that the approach used with the ISOCS system was one of verification of the presence of material. They have purposefully implemented a conservative quantification algorithm and anticipate a better understanding of the facility holdup as they gain more experience.

Table 2. Normalized uranium mass results for a vacuum line.

Group	Date	Normalized Uranium Mass	Random 1σ Uncertainty in Normalized Uranium Mass
GGH-A	July 2004	1.00	0.21
GGH-B	July 2004	0.87	0.17
GGH-C	July 2004	0.73	0.43
GGH-A	October 2004	0.75	0.05
GGH-B	October 2004	0.96	0.06
ISOCS-A	September 2004	0.48	none reported
ISOCS-B	September 2003	0.57	none reported
ISOCS-C	September 2004	0.38	none reported

Because the deposits were greater than the limiting thickness to the 186-keV gamma ray at many measurement points along this vacuum line, masses reported using the GGH method are determined from 1001-keV data. The average correction for self-attenuation was 10 – 15% for the five sets of data, and other corrections are considerably less. All the masses reported from the GGH measurements agree with one another at the 2σ level for random uncertainty, and four of the five agree at the 1σ level with the average of all five measurements being 0.86 ± 0.10 where the uncertainty is the propagated random uncertainty in the mean. The 1σ standard deviation of the five GGH results is 0.12. The average of the ISOCS results is 0.48 with a standard deviation of 0.09 for the three measurements. The ISOCS measurements were typically a few points along the vacuum line, but for many hundreds of seconds each, thus, the random uncertainties are negligible compared to the systematic uncertainties.

Plotting the normalized specific mass as a function of measured distance or linear position along the process equipment can be useful. Such maps reveal where holdup accumulates faster relative to other points along a line. The user can also compare equivalent maps over time to and possibly predict when and where clean-outs will be required. Figure 2 shows the specific mass as a function of distance for this vacuum line as measured by group GGH-A in July 2004. Note larger error bars at locations of thicker deposits caused by blow-up of the relative uncertainty propagated through the self-attenuation correction algorithm.

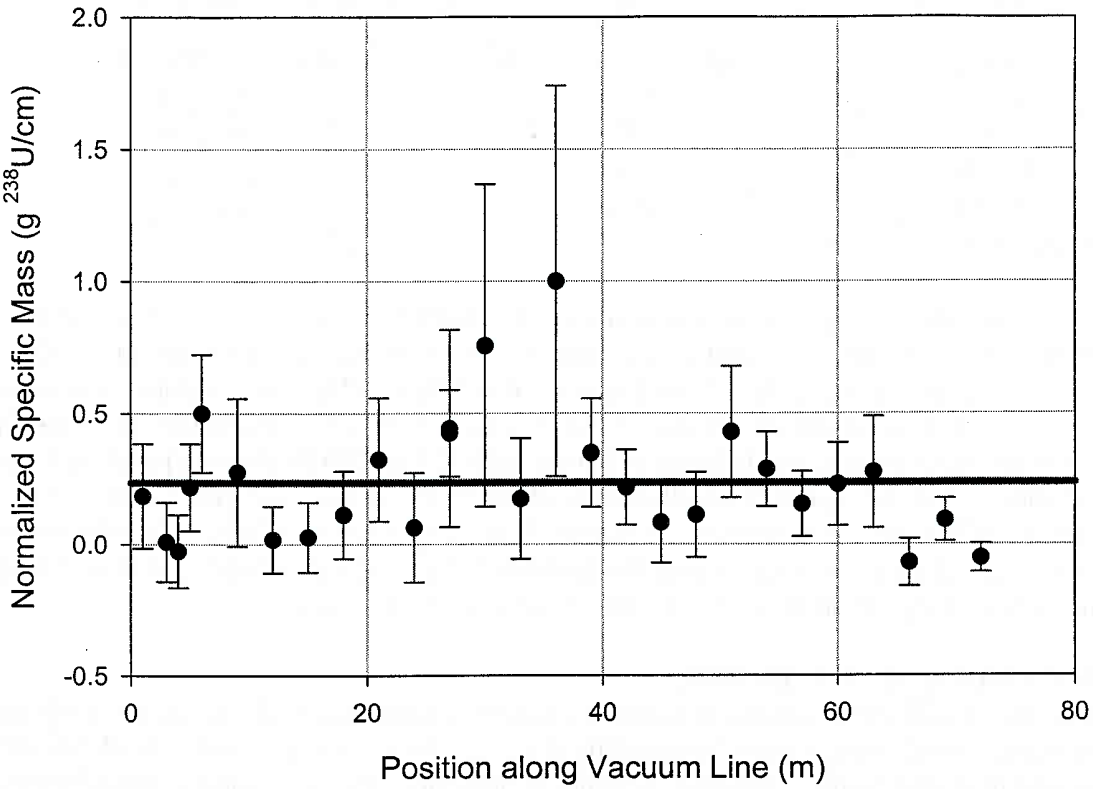

Figure 2. Normalized specific mass of ^{238}U vs. position. The red line indicates the average normalized specific mass (0.236 g/cm).

MEASUREMENT OF A MOCK STANDARD

As seen in Table 2, the results from GGH and ISOCS measurements do not agree well. The two methods generally differ by a factor of two on this and other equipment that was measured. An attempt was made to validate the two quantitative measurement methods by creating a standard that resembles actual deposits measured by both GGH and ISOCS. The facility distributed equal amounts of uranium oxide feed material into 10 glass jars of 8-cm diameter. These jars were then arranged on the floor end-to-end to form a two-meter-long line "deposit" with a known inventory of 3500 g of uranium. The simulated deposit closely resembles the actual material geometry and average specific mass in the vacuum line described in the previous section. This simulated deposit was measured by two teams using separate equipment and the GGH method, and by third team using the ISOCS equipment and method. The GGH measurements were performed, as they are done on the process equipment, at multiple locations along the length of the extended equipment. The 1001-keV gamma ray data were used in the analysis. The team using the ISOCS method made two measurements of the "deposit" in two different measurement geometries, each viewing and counting the entire 2-m-long standard in a single measurement. Typically, ISOCS measurements of extended process equipment (ducts or pipes) rarely, if ever, view the entire segment of equipment in a single count, and so the measurement geometry for the 2-m-long standard differed from the

geometric methodology of the ISOCS setup for measurements of process deposits. The results of this exercise are shown in Table 3.

Table 3. Comparison of results for the 3500-g, 2-m-long uranium "standard"

Group	Measured Uranium (g)	Random uncertainty (g)
ISOCS1	3000	None provided
ISOCS2	3000	None provided
GGHa*	3260	520
GGHb*	4040	560

* Results are based on 1001-keV data.

The finite-source and the average self-attenuation corrections for this geometry were 1.057 and 1.099 respectively for the 1001-keV gamma-ray. The two GGH based measurements agree with each other and with the reference value of the 2-m-long standard to within 1σ (random uncertainty). Because no uncertainty is reported for the ISOCS value, it is difficult to ascertain the "goodness" of the measured mass. Nonetheless, the differences between ISOCS and GGH do not appear as large as those determined from the repeated measurements of the 74-m vacuum line (see Table 2). The possibility that ISOCS treats the geometry of a standard correctly when it is fully within the field of view of the detector but fails to correctly treat the geometry of a more extended deposit (such as a long duct many times larger than the field of view) should be investigated.

ANALYSIS OF SYSTEMATIC ERROR

A drawback to the ISOCS method is the absence of a reported uncertainty. If one used the reported activity uncertainty, which comes from the counting statistics, the uncertainty in mass determined by ISOCS would be grossly underestimated. In order to determine the magnitude of the effect that the uncertainty in the unknowns have on the final mass result it is necessary for the user to re-analyze the measurement by adjusting the user provided measurement parameters with the largest contribution to the systematic measurement uncertainty. An example is that the user must provide the ISOCS software with the thickness of the deposit, which is very difficult to know accurately, but an uncertainty in the thickness of ±50% may be reasonable. The user would then analyze the measurement with a deposit thickness that varies by ±50% to determine the systematic uncertainty in the mass due to the uncertainty in the deposit thickness. This should be repeated for other measurement parameters for which uncertainties are significant and can be estimated. The final uncertainty in the mass would be a sum in quadrature of random (counting statistics) and systematic uncertainties. This numerical approach to estimation of systematic uncertainty also applies to and is used in the GGH analysis of uncertainty. However, counting statistics dominates the GGH uncertainty for measurements of the standard at 1001-keV in short count times.

CONCLUSIONS

Using low-resolution, portable, room temperature gamma-ray spectroscopy systems and GGH analysis tools, the uranium holdup was measured in the ventilation and vacuum lines of a LEU fuel-fabrication plant. Repeated measurements of holdup deposits show good agreement.

These measurements are the first to achieve or demonstrate the following.

- Low-resolution quantitative gamma spectroscopy of holdup in a wide dynamic range (100 –1000-keV).

- Highly portable quantitative measurements of very thick deposits of uranium oxide.
- Practicality of measuring 1001-keV gammas from a large holdup deposit in a "sea" of highly penetrating 1001-keV gammas from room background.
- Viability of the portable quantitative measurements for LEU holdup on the scale of a large, complex facility.
- Implementation of the generalized holdup correction algorithms for LEU.
- Broad understanding of the range of self-attenuation effects in an LEU-solids plant, and the assurance that the gamma techniques are suited to this range.
- Assurance of the accessibility of most deposits to measurements performed using highly portable gamma spectrometer detectors.

The measurements reinforce the need for the following procedures and capabilities – many unique to LEU measurements – for successful routine implementation of portable low-resolution gamma-ray measurements:

- A spectrometer detector with good detection efficiency through 1001-keV.
- Simultaneous measurements of both 186- and 1001-keV gammas.
- A fixed internal reference source of ^{241}Am for measurement control spectrum-by-spectrum and gain-drift compensation.
- Alternative to ^{238}U source for timely measurements of off-axis detector response during calibration.
- A measurement, with the tungsten plug inserted, of room background for every deposit measurement.
- Conservative setting of the ROI on the 186-keV peak to avoid the effects of interference from the 238-keV gamma.
- Full automation of the data acquisition and measurement control. This will result in a measurement time savings of an order of magnitude [4].
- Stabilization against or compensation for scintillator gain drift.
- Alternative geometric models for self-attenuation.
- Implementation of (an energy-dependent) mean gamma interaction depth in the crystal.
- Full access by users to all input parameters used by the analysis tool to assure ability for numerical determination of systematic effects.
- Implementation of numerical evaluation of systematic contributions to measurement uncertainty.
- Flexible implementation of analysis for either (186- or 1001-keV) depending on equipment cleanout status.
- Implementation of the 3σ warning in approach to thick deposits.
- RFID tagging and readout of holdup measurement locations.

ACKNOWLEDGEMENTS

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