Holdup Measurement System 4 (HMS4) - Automation & Improved Accuracy

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Abstract

MEGA/RAM* (Measure & Evaluate Generalized-geometry γ-ray Assay / Rapid Automated Method) is a rapid automated approach – based on portable γ-ray spectroscopy – for quantifying uranium and plutonium deposits *in situ* for holdup-accountability and criticality safety. The project is funded by the Department of Energy Office of Security Policy, with additional funding support from Y-12, as a cooperative effort between the Oak Ridge National Laboratory, Y-12 National Security Complex and Los Alamos National Laboratory. The software and hardware system developed by this project is called the "Holdup Measurement System 4 (HMS4)." The system implements new commercial multichannel analyzers (MCAs), portable field controllers, and software platforms. New generalized corrections for finite-source dimensions and γ-ray self-attenuation are used to achieve improved accuracy. HMS4 employs a new quantitative deposit screening for criticality safety and also automates and enforces uniform measurement control in the field. Using HMS4 for measuring routine plant-wide holdup reduces costs of these safeguards measurements by making them almost a one-person task. A single user could measure and analyze hundreds of deposit-locations per shift. The Los Alamos Plutonium Facility and the Y-12 National Security Complex are hosting in-plant tests of the prototype HMS4 applied to plutonium and uranium, respectively. Both these facilities currently use ten-year-old technology, which HMS4 replaces, for automated measurements of holdup deposits. Transfer of the new automation technology to the commercial source of the portable MCAs is underway.

Introduction

Gamma-ray spectroscopy is an important technique for the measurement of nuclear material holdup and has several advantages over neutron techniques. Because γ rays can be collimated relatively easily, the locations and distributions of deposits can be defined by these measurements. Shielding can also minimize room background radiation, which can be as significant a contributor of γ rays as the holdup itself. Another advantage of γ rays is that multiple isotopes and elements can be measured independently and simultaneously by choosing the detector and peaks appropriately. Lastly, γ-ray detectors and the required electronics can be small, light in weight, portable, and relatively inexpensive compared to neutron systems, such as the Neutron Slab Detector.

Nevertheless, γ rays are easily attenuated (unlike neutrons), and the attenuation can be quite significant. Attenuation by the equipment walls and other shielding can be readily calculated, but self-attenuation by the nuclear material can be much more difficult to determine. If self-attenuation is ignored, the measured quantity will have a negative bias. Another major contributor to the accuracy of γ-ray holdup measurements comes from the geometric model used to quantify the holdup. The model itself gives rise to a systematic negative bias in the assay result because the finite geometry of any real deposit does not conform strictly to the geometric model. The finite-source effect will be described in more detail later in this paper.

There is also a need, beyond quantitative holdup measurements, to screen deposits in process equipment radiologically to detect the presence of unsafe quantities. Rapid screening is typically accomplished by verifying that the γ-ray count rate (dosimetric) is less than a predetermined limit. However, spectral variations or equipment attenuation alone can reduce a measured dose rate. Self-attenuation can cause additional order-of-magnitude reductions in the emitted γ rays from large deposits. Therefore, present systems use arbitrarily and conservatively low limits in screening for criticality safety. An automated Holdup Measurement System, HMS[1] was designed in 1991 to monitor hundreds to thousands of possible holdup areas that represent criticality safety concerns. Improving the attenuation calculations for this screening process by using the same model developed to quantify holdup will promote the use of more realistic limits in this process.

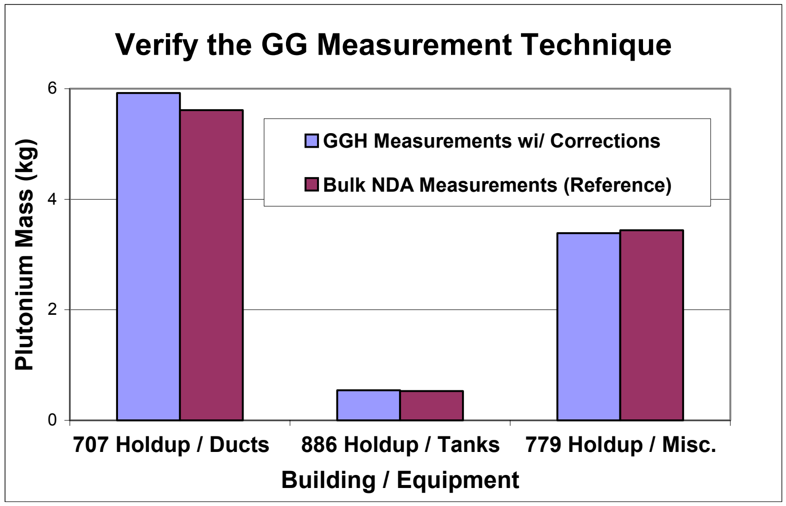
This quantification model is the Generalized Geometry (GG) Approach to γ-ray holdup measurements, which is documented elsewhere[2]. The methodology was incorporated in the first semi-quantitative automated Holdup Measurement System, HMS2[3], followed quickly by the next version, HMS3[4]. HMS3 was developed for use by DOE facilities to enable thousands of measurements to be performed plant-wide during routine inventory periods. The project was carried out jointly by Los Alamos (under DOE OSS support) and Oak Ridge (with support from Y-12), beginning in 1996. The system automates hardware setup; calibration; measurement control; data acquisition and storage; data reduction and analysis using the GG holdup algorithms; correction for room background and equipment attenuation; computation of holdup in equipment, areas and facility-wide; and statistical-error propagation. It uses bar codes to identify the holdup measurement locations; and a database to store information on the equipment and measurement for each location. Users of the automated system set up to measure the barcoded locations can obtain thousands of measurement results, the total holdup mass and its distribution in the facility during the inventory week. The equivalent measurements performed manually are impractical, requiring many times the effort, as demonstrated in published comparisons[5].

New Corrections

The MEGA/RAM project, sponsored by the Department of Energy Office of Security Policy, has produced a new, rapid, automated system for determining accurate holdup quantities plant-wide and will allow for semi-quantitative screening of deposits for criticality safety. The result of this project is the "Holdup Measurement System 4 (HMS4)". Measurements of holdup are required for accountability, yet plant-wide holdup measurements for inventory are not practical without automation. Automation does not eliminate the need for users to perform portable γ-ray spectroscopy measurements, but it decreases the influence of user-subjectivity on the NDA result and enforces measurement control. Additionally, new analysis approaches have been shown to improve the accuracy of γ-ray holdup measurements[6]. The improved holdup measurements are consequently better suited than ever for accountability of nuclear materials. Built on the generalized-geometry (GG) approach to γ-ray holdup measurements, the methodology corrects for finite source dimensions and self-attenuation using the new, general techniques developed recently for the GG measurements. It automates measurements using the newest portable MCA technologies, but is also backward compatible to presently implemented hardware. The new system is being implemented in two DOE facilities and is also being commercialized.

Reference 6 derives the systematics of a new empirical parameter, the width of the deposit, to determine the finite-source (1) and the self-attenuation (2) corrections from the measured data. 1) The collimated detector response peaks at the center of the detector field of view. A point (or line) source in the GG model is assumed to have no width. Such a source positioned at the center and measured with the collimated detector gives the maximum response. Actual point (or line) deposits have finite width, which biases the GG result low. A best estimate of this width (possibly based on equipment dimensions) is the basis of a correction that uses the known dependence of the detector response on displacement from the center. Overestimated widths overestimate the finite-source correction, and vice *versa*. 2) Self-attenuation is also calculated using this width parameter, which converts the measured mass of a point (or linear density of a line) to the areal density, required to quantify self-attenuation. The same overestimated width underestimates true areal density, which underestimates the self-attenuation effect, and *vice versa*. The effect of over- or under-estimating deposit width is opposite in direction for self-attenuation and finite-source corrections. Therefore, the corrected measurement result is less sensitive to uncertainty in the empirical width parameter. This is an added strength of the new correction factors and a compelling reason to employ both types of corrections.

Figure 1: Verification of HMS4 and GG Technique[7]



Recent GG holdup measurements used facility-wide with the new, generalized corrections for finite-source effects and γ-ray self-attenuation agree with reference values determined by NDA measurements of bulk materials from controlled cleanouts of these facilities. Figure 1 shows preliminary results of this study.

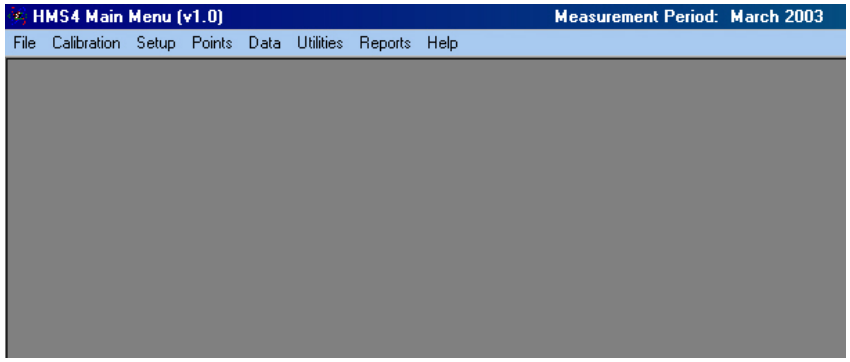
Software Development

HMS4 includes two sets of programs: the main program runs on a host personal computer (PC), and the "field" programs run on a bar-code reader or portable PC. The bar-code reader or portable PC is often referred to as a controller. The main host computer program performs setup and calibration of multichannel-analyzer/detector pairs, loads the controllers with operational parameters, receives measurement data from the controllers, maintains measurements and derived results in databases, and prints reports. The field controller programs set up the multichannel analyzer (MCA), control data acquisition, automate measurement control, store measurement data as accumulated, and allow the user to review previous collected data and spectra. Four other host computer programs, which are included in the distribution, are as follows: 1.) The stand-alone Windows-based Controller program (HMS4 Controller), 2.) A stand-alone MCA control selection program (MCA Switch 2), 3.) An upgrade program (HMS4mdb) to be used to update existing older HMS3 databases to HMS4 format, and 4.) A program (Spectra Split) for extracting the embedded region-of-interest (ROI) information from a saved controller spectrum.

HMS4 is a Microsoft Windows-based software package that has evolved from three earlier, sequential program versions. HMSII, the first in the series, was a DOS based program written in Microsoft FoxPro[@]. The second, HMS3 v1.0, was based on Microsoft Windows and written in Microsoft Visual Basic 4.0. The third, HMS3 v2.0, was written in Microsoft Visual Basic 6.0; the functionality was the same has HMS3 v1.0, but a newer compiler made the step to the next compiler a little easier. HMS4 is written in Microsoft Visual Basic .NET, which is part of the Microsoft Visual Studio .NET 2003 development package. HMS4 uses Microsoft Access (Microsoft Office 2000/XP format) database files. The reports are generated with the Crystal Decisions, Inc., Crystal Reports report generator, which is included with the Visual Basic .NET package. The software for the controllers, as previously mentioned, takes two forms. The software for the bar-code reader controllers is written in the Microsoft eMbedded Visual Basic (part of Microsoft eMbedded Visual Tools v3.0) development package for Windows CE. This software will run on any Microsoft Pocket PC operating system device. The software written for the PC or Laptop Controllers is written in Microsoft Visual Basic .NET.

The Windows-based HMS4 provides the user with several enhancements over the previous HMS3 versions. It offers a completely new menu-based environment. Figure 2 shows a portion of the main screen of HMS4. The menu bar dropdown boxes allows access to the various system functions.

Figure 2: HMS4 Main Menu

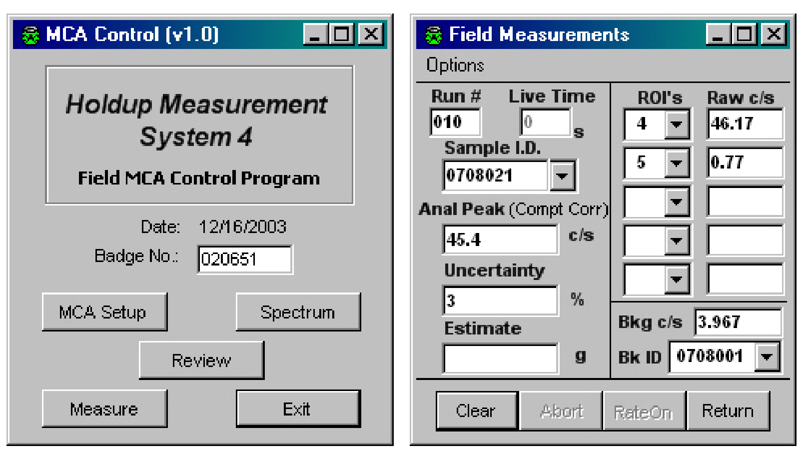


The analysis software contains several new holdup correction algorithms such as the finite-source correction and the self-attenuation correction, for the capability of full error correction. HMS4 now supports twenty (20) spectral regions-of-interest (ROIs) to particularly aid the user measuring plutonium. The data from each measurement period (or campaign) can now be easily accessed from the main menu. All measurement data dumps are date and time-stamped and allow for an 80-character comment field, which can be used for extra notes. Serial communications between the host computer or the bar code reader/controllers and the various MCAs have been improved using DLLs designed by the MCA distributor. This has helped to eliminate timing and handshaking problems inherent in the earlier HMS3 versions. Many improvements have been made in the way that HMS4 performs background calculations, and now, the user has the flexibility to make 20 consecutive background measurements and reference these to particular measurements of deposits.

Hardware Support

HMS4 supports four modern multichannel analyzers (MCAs) [8]. The field controller software of HMS4 also supports several bar code readers from multiple manufacturers, the main requirement being the operating system, Pocket PC (2000, 2002 or 2003). Several monochrome and color versions have been satisfactorily tested. If a barcode reading device is not really necessary, any Pocket PC based device such as a Personal Data Assistant (PDA) can run the controller software. As earlier mentioned, there is also a full Microsoft Windows version of the controller software for use of a laptop computer in the field. Figure 3 shows the main screen for the controller program and the screen for the acquisition window, the heart of the controller program.

Figure 3: Controller Main Menu and Field Measurement



The γ-ray detector types supported by HMS4 are those supported by the MCA chosen. Detectors tested include sodium iodide (NaI), large cadmium zinc telluride (CdZnTe), and germanium (Ge). Other possibilities exist as well. The detector must have a radioactive reference source attached in order for all of the integrated measurement control features of HMS4 to work fully. A source of

^{241}Am (~1 μCi) is a common example.

The algorithms and methods for MEGA/RAM have been developed, specified, verified and documented by Los Alamos, consistent with measurement needs for uranium and plutonium facilities. Oak Ridge has developed and is commercializing the automation software. The facility testing has been the responsibility of both Los Alamos and Oak Ridge. Facility tests have been conducted alongside the current efforts for holdup/criticality-safety measurements, and in conjunction with planned programs to train users at their respective facilities. In-plant evaluations of HMS4 are being conducted at the Los Alamos National Laboratory and at the Y-12 National Security Complex. The evaluation at the Los Alamos Plutonium Facility is supported by the Safeguards and Security group in the Nuclear Materials Technology division. The Y-12 evaluation is supported by the NDA group of the Analytical Chemistry Organization. Both of these potential users of HMS4 are current users of the current HMS3 system for automated measurements of holdup. Both use NaI detectors and large CdZnTe detectors for holdup measurements and are implementing the newest portable MCAs. Both facilities, along with Hanford, Portsmouth and Rocky Flats, now manually implement the new corrections for finite-source and self-attenuation effects. All four facilities plus Savannah River, Hanford, Irwin and Paducah use the GG approach to calibrate and measure holdup. Most also use dosimetry for criticality-safety screening. Therefore, all of these facilities are potential users of the HMS4.

Testing HMS4 and the Automated System

Laboratory testing of the automated system was performed from data collected from students at a holdup class entitled “Automation of Uranium Holdup" held in Oak Ridge in March, 2004 for 16 people from Y-12. Each of four four-person teams calibrated a separate MEGA/RAM system consisting of a sodium iodide detector and portable MCA, and then performed measurements on each of six simulated holdup setups loaded with uranium working reference materials. The automation software is HMS4. The geometric distributions of the reference materials inside the simulated process equipment were representative of point, line and area deposits of ^{235}U in the plant. Finite-source corrections for the simulated deposits can be 20-30%, depending on measurement distance. Self-attenuation is small for some but not all of these reference materials.

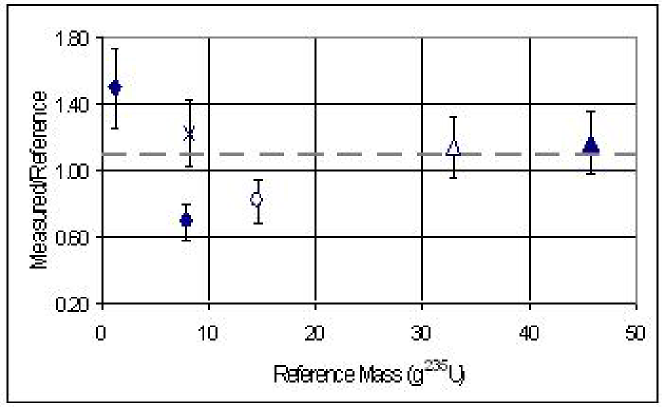
Table 1 describes the six simulated holdup setups, and gives the reference value for the ^{235}U loading of each. Figure 4 plots the mean ratio of the (MEGA/RAM-) measured ^{235}U loading to the reference loading obtained for the measurements of each setup by the four groups vs. the reference loading. The error bars indicate the standard deviation in the mean of the four measurements and are comparable to statistical uncertainties. This indicates that other random effects do not contribute substantially.

The MEGA/RAM measurements of uranium materials are in good agreement with the reference values with two exceptions, which are expected. 1) The reference items in the Small Round Duct are composed of lower-enriched “recycle” material with the characteristic 238-keV γ-ray interference in the continuum-background region of the 186-keV assay peak. Ordinarily, Y-12 performs measurements of this type of material with the large CdZnTe detectors that are considerably less susceptible to this bias because of improved energy resolution compared to NaI detectors. 2) The reference materials in the Pipe Array include several items that are nearly infinitely thick to 186-keV γ rays. The ability to quantify such materials breaks down for very thick samples (also refer to the results below obtained using plutonium standards), which are definitely not representative of most holdup deposits.

Table 1: Holdup Class Setups

Equipment	Ref g ^{235}U	Symbol
HEPA House	1.2	◆
Small Round Duct	7.9	●
Small "S" Duct	8.2	X
Pipe Array	14.7	○
Large Round Duct	32.9	△
Large "L" Duct	45.7	▲

Figure 4: Result of Holdup Measurements for the Class



Additional laboratory tests of the automated system using plutonium were performed at Los Alamos:

- Compare reduced spectral data (net peak count rates and uncertainties) with hand calculations using the appropriate algorithms.
- Calibrate generalized-geometry holdup measurements. Compare calibration parameters and uncertainties with hand calculations using the appropriate algorithms.
- Analyze simulated plutonium holdup to give specific masses and uncertainties. Compare with 1) hand calculations using the appropriate algorithms, and 2) reference values for the specific masses of the plutonium standards used to simulate holdup.

The tests used measurement systems composed of large CdZnTe detectors (15 × 15 × 15 mm^3), and portable DSP multichannel analyzers operating under PC control. An encapsulated plutonium metal foil standard was used for the calibration measurements. Three encapsulated plutonium oxide standards were used, along with the metal calibration foil, to simulate point holdup deposits of different thickness.

Columns 2 and 3 of Table 2 are the mass and areal density data for the plutonium metal calibration standard and the three plutonium oxide standards. Columns 4-6 are the multiplicative corrections for container attenuation, the finite-source effect, and self-attenuation, all calculated by the HMS4 software.

Table 2. Plutonium Metal (C) and Oxide (A) Standards (94% ^{239}Pu)

ID	g ^{239}Pu	g Pu/cm^2	CF_{EQ}	CF_{FINSRC}	CF_{SELF}	Ratio	% 1-σ_{Ratio}
C1	1.71	0.579	1.134	1.007	1.086	1.07	6%
A1	1.88	0.355	1.196	1.011	1.047	1.05	11%
A2	4.67	0.881	1.196	1.011	1.115	1.03	8%
A3	9.34	1.762	1.196	1.012	1.214	0.95	7%

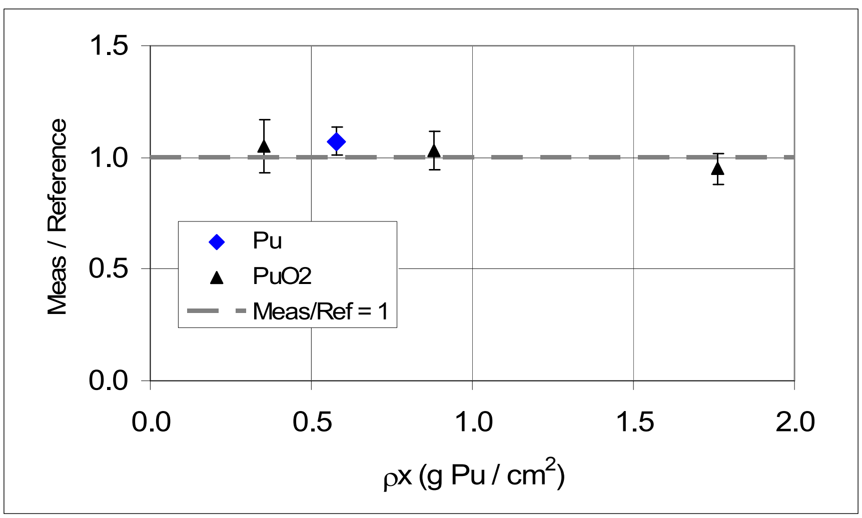
The correction for container (or "equipment") attenuation, column 4 of Table 2, is determined using parameters derived from composition and thickness information entered into the HMS4 database that describes each holdup measurement point. The steel (cladding) thicknesses varied from 1.8 to 2.5 mm for the plutonium metal and oxide standards. The corresponding attenuation effects were 13% and 20%, respectively. Ignoring equipment attenuation effects would result in a negative bias in the measurement results. These effects are traditionally included in analysis procedures for holdup measurements because process equipment dimensions are usually known.

The finite-source correction, column 5 of Table 2, is determined using the empirical width (typically the inner diameter of the pipe or other cavity that contains the holdup) of the standards, which was stored in the HMS4 database for each measurement. The finite-source effects are small – approximately 1% – in this case because the 2-to-2.5-cm-diameter plutonium metal and oxide standards were positioned at relatively large distances (40 to 43 cm) from the detector for these measurements. Corrections for finite-source effects have not been included in analysis procedures for holdup measurements traditionally, although these can be large effects in some cases and always contribute to negative bias.

The self-attenuation correction, column 6 of Table 2, is also determined – using the same deposit width parameter that determined the finite-source correction – to convert the measured specific mass (mass for point deposits; mass per unit length for line deposits) to measured areal density, ρx_{MEAS} (mass per unit area). A far-field attenuation form converts the measured areal density of point, line and area deposits to ρx, the measured areal density corrected for self-attenuation. Columns 7 and 8 in Table 2 give the ratio of measured ρx to the reference value (column 3 of Table 2) and the 1-σ uncertainty in the ratio propagated from counting statistics.

Figure 5 is a plot of ratio of the measured ρx (or mass) to the reference ρx (or mass) of ^{239}Pu vs. the areal density of the measured standards. The error bars indicate the counting statistics. Agreement between measured and reference values is within statistics, with a mean ratio of 1.03 ± 0.03 (1σ). The possible trend toward a negative bias as the oxide sample thickness increases could be a result of limitations of a far-field geometry assumption for the self-attenuation correction as the sample gets very thick – in this case more than an order of magnitude thicker than the thickness of most holdup deposits.

Figure 5: Results of the ^{239}Pu measurements.



The self-attenuation correction is ~5% for the smallest oxide standard. The plutonium areal density for this source is 0.355 and 0.579 g/cm^2, respectively. The thickness of holdup deposits (the typical range is 0.01-0.3 g actinide/cm^2) rarely exceeds 0.3 g of actinide per cm^2, so the two smallest standards are near and above the upper end of the range for typical deposits. The metal standard and the two larger oxide standards (0.579, 0.881 and 1.762 g Pu/cm^2) are, respectively, two, three and six times thicker than the upper end of the thickness range for expected holdup deposits. The corresponding self-attenuation corrections are approximately 9%, 12% and 22%. Ignoring the correction causes a negative bias in the measurement result in all cases. Corrections for self-attenuation effects have not been included in analysis procedures for holdup measurements traditionally.

The results of the hand calculations using the appropriate algorithms for data reduction, data analysis and error propagation agree with the corresponding results determined by the HMS4 software. This validates the programming of algorithms for reduction, analysis and error propagation for the measurement data.

Summary of Results

The Accurate MEGA/RAM's new automation software for holdup measurements, HMS4, will extend significant benefits to all facilities with routine holdup measurement needs because of three new, major capabilities. 1) Accuracy: Currently HMS3 lacks the correction algorithms for finite-source and self-attenuation effects. These two effects can cause γ-ray holdup results throughout the plant to be biased low, which is not acceptable for accountability. The new, recently documented correction algorithms used in the HMS4 software are both consistent with the GG method and are mutually consistent. They are nearly transparent to the user when implemented in an automated system but very cumbersome and time consuming to implement manually. 2.) Compatibility: Currently, HMS3, which must automate the setup and operation of the MCA, is compatible with only four commercial portable MCAs: three are 5-to-10-year-old technologies and one is 20 years old. (One of these MCAs can only be implemented with NaI. Both Los Alamos and Y-12 are performing holdup measurements with the large CdZnTe detectors.) Several new, smaller, commercial, portable MCAs with better performance (and some that include Digital Signal Processing [DSP]) cannot be used with the current HMS3 software. 3.) Safe-Deposit-Thickness Readout: Facilities that perform holdup measurements also use passive γ-ray measurements to screen deposits for criticality safety. With adaptation of the new correction algorithms used in the HMS4 software, a quantitative safe limit for deposit thickness can be confirmed automatically. Without an equivalent approach, criticality-safety screening using passive γ rays is non-quantitative, arbitrary and necessarily conservative as a result. The HMS4 is needed for accuracy of automated holdup measurement results, compatibility with current commercial technology and capability to provide a quantitative result that relates directly to criticality safety.

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