This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-96SR18500 with the U.S. Department of Energy.

HOLDUP MEASUREMENTS FOR VISUAL EXAMINATION GLOVEBOXES AT THE SAVANNAH RIVER SITE

R.A. Sigg, V.R. Casella, R.A. Dewberry and F.S. Moore
Savannah River National Laboratory
Aiken, SC 29808

Visual Examination (VE) gloveboxes are used at the Savannah River Site (SRS) to remediate transuranic waste (TRU) drums. Noncompliant items are removed before the drums undergo further characterization in preparation for shipment to the Waste Isolation Pilot Plant (WIPP). Maintaining the flow of drums through the remediation process is critical to the program's seven-days-per-week operation. Conservative assumptions are used to ensure that glovebox contamination from this continual operation is below acceptable limits. Holdup measurements are performed in order to confirm that these assumptions are conservative. High Cs-137 backgrounds in the VE glovebox areas preclude the use of a sodium iodide spectrometer, so a high-purity germanium (HPGe) detector, having superior resolution, is used.

Plutonium-239 is usually the nuclide of interest; however, Pu-241, Np-237 (including its daughter Pa-233) and Pu-238 (if detected) are typically assayed. Cs-137 and Co-60 may also be detected but are not reported since they do not contribute to the Pu-239 Fissile Gram Equivalent or Pu-239 Equivalent Curies. HEPA filters, drums and waste boxes are also assayed by the same methodology. If – for example - the HEPA is contained in a stainless steel housing, attenuation corrections must be applied for both the filter and the housing. Dimensions, detector locations, materials and densities are provided as inputs to Ortec's ISOTOPIC software to estimate attenuation and geometry corrections for the measurement positions. This paper discusses the methodology, results and limitations of these measurements for different VE glovebox configurations.

INTRODUCTION

Department of Energy's Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico places limits on the waste that can be shipped to the facility. The facility's Waste Acceptance Criteria (WAC) ensures that waste forms are stable and that these forms can be safely managed. Therefore, the WAC excludes certain wastes forms that contain liquid waste, pyrophoric materials, corrosive materials, ignitable waste, and compressed gases. The TRU waste must also contain 100 nanocuries or more of transuranic elements per gram.

Sites that ship waste to the WIPP must confirm that the shipment does not include any these noncompliant waste forms. The Savannah River Site meets these requirements through initial measurements that determine TRU activity levels and that identify noncompliant items. SRS organizations are using several glove box facilities for remediating contact handled drums that contain noncompliant items. When noncompliant items are found, personnel at these facilities open the drums, remove and/or stabilize noncompliant materials, and repackage the drums.

Maintaining the flow of drums through the remediation process is critical to the program's seven-days-per-week operation. Residual fissile materials in the glovebox trains are assayed to assure that the materials remain below levels that are defined in Technical Safety Requirements (TSR). Operations personnel assume that 10% of the fissile material is held up in the glovebox until assays, performed by Savannah River National Laboratory (SRNL) personnel, conservatively determine maximum values for the holdup. Schedules for the assays vary widely due to differences in fissile material contents of the drums and drum throughput.

SRNL personnel routinely apply high purity germanium (HPGe) gamma-ray nondestructive assay techniques to determine the radioactive material contents in numerous other items. Examples include objects from facilities that are being decontaminated and decomissioned, excess nuclear materials under safeguards, waste items in 55-gallon drums, heat exchangers, and B-25 waste boxes.

TECHNIQUE

Compton scattered backgrounds from Cs-137 in the VE glovebox areas preclude use of low resolution gamma-ray spectrometers for lower energy gamma rays, such as those from Np-237/Pa-233, Pu-238 and Pu-239, the radionuclides of primary interest. Therefore, SRNL uses high purity germanium (HPGe) detectors for these holdup measurements. Figure XYZ, a typical HPGe spectrum acquired at one of the facilities, shows the presence of Pu-239 in the presence of significant background that is primarily due to Cs-137.

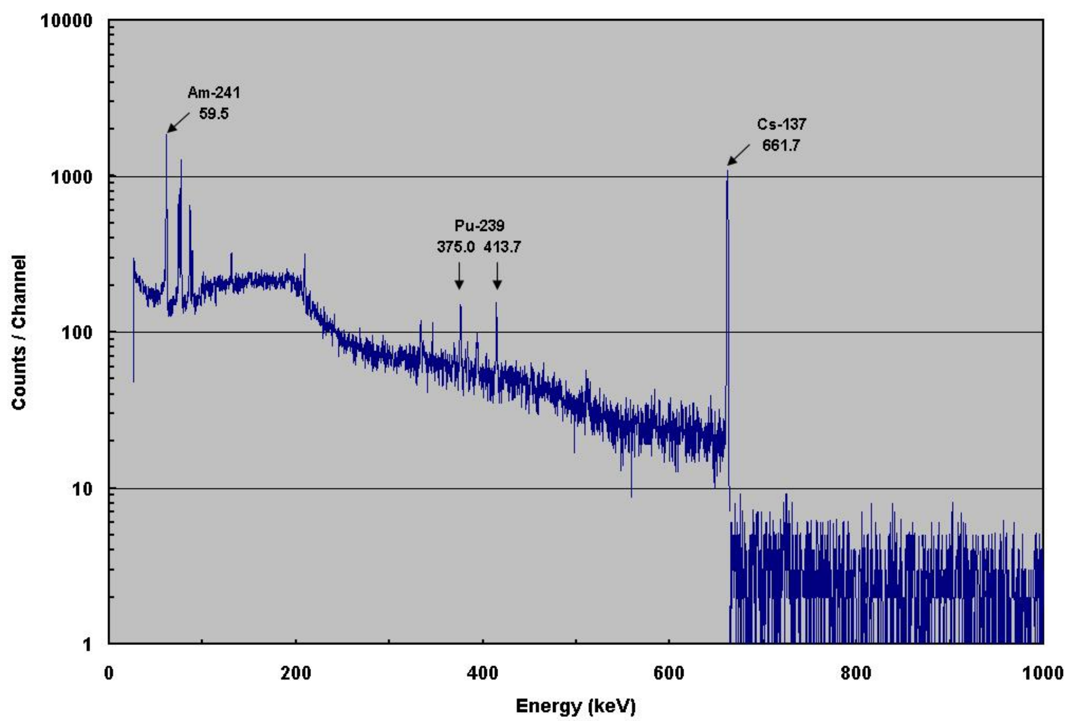

Figure 1. An HPGe gamma-ray spectrum at a VE glovebox section showing Pu-239 in the presence of a high Cs-137 background.

Calibration

The efficiency calibration of HPGe detectors generally follows one of two separate approaches, both of which are approved by the American National Standards Institute (Ref. 1). The more general calibration approach determines of full-energy peak efficiencies as a function of gamma-ray energy. In the other approach, standardization coefficients are determined by directly comparing results to a standard source of the same radionuclide.

We employ the more general method since it enables quantification of all radionuclides that are observed in a gamma-ray spectrum rather than limiting the analysis to only one or two calibrated radionuclides. Calibration data are acquired with NIST-traceable point-source standards, an Analytics, Inc. mixed-gamma standard and an Isotope Products Laboratory Eu-152 standard. The standards are positioned on an on-axis position 12 inches from the face of each high purity germanium detector. Canberra Inspector electronics are used with Ortec portable HPGe detectors to acquire the spectra. Peak energy, area and efficiency data from spectra analyzed by Canberra's Genie 2000 software (Ref. 2) are saved to files for use by Ortec's ISOTOPIC software (Ref. 3).

Geometry and Attenuation Corrections

Ortec's ISOTOPIC software applies gamma-ray emission intensity, geometry and attenuation corrections to Genie 2000's peak area and efficiency data to determine nuclear material contents in assayed items.

Geometry correction factors permit extending point-source calibrations to assays of objects having other shapes. While the ISOTOPIC program comes with several predefined descriptions of waste containers, it provides a relatively simple user interface for describing other objects. We have applied the method to describe geometry corrections for holdup measurements on glovebox trains for VE, MRS and stabilization.

ISOTOPIC also determines attenuation corrections from knowledge of the container materials, matrix composition and density, and percent composition of high atomic number materials (U or Pu). A unique and important feature of ISOTOPIC is that it provides a graphical display of these results for each gamma-ray from a radionuclide (e.g**.** Figure 2).

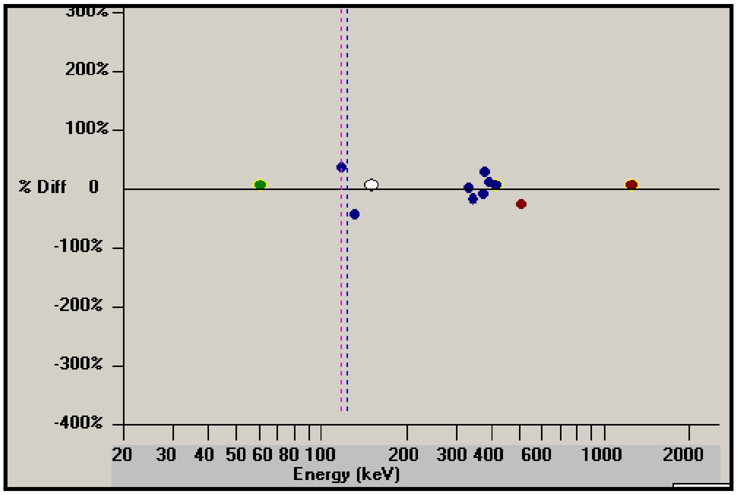

Figure 2. ISOTOPIC's plot of output values as a function of gamma-ray energy. The points shown in blue are for various energies characteristic of Pu-239.

This visual interface is useful in determining whether the attenuation corrections that are applied in calculating radionuclide activities are appropriate. Since many radionuclides emit gamma-rays at several energies, analysis of the data should give the same value for the number of Curies or grams of nuclear material in the object. That is, each gamma-ray should give nearly the same answer (within uncertainties) for the activity or mass of the radionuclide. ISOTOPIC allows adjusting the attenuation parameters until the results match this physical reality.

Performance Assessments

We tested ISOTOPIC's performance by comparing its results to

- Well-known standards for several measurement geometries, some having significant attenuation.
- Geometry and attenuation corrections to those provided by MicroShield (Ref. 4), a point-kernel shielding computer code.

HEPA filters for two of SRS's glovebox trains can only be assayed using gamma-rays that penetrate the filters' stainless steel shells and their thicker stainless steel housings. Gamma-ray spectra using the calibration standards with and without the filter and its housing, clearly show significant attenuation in low energy regions important for analysis Np-237/Pa-233, and Pu238, and Pu-239.

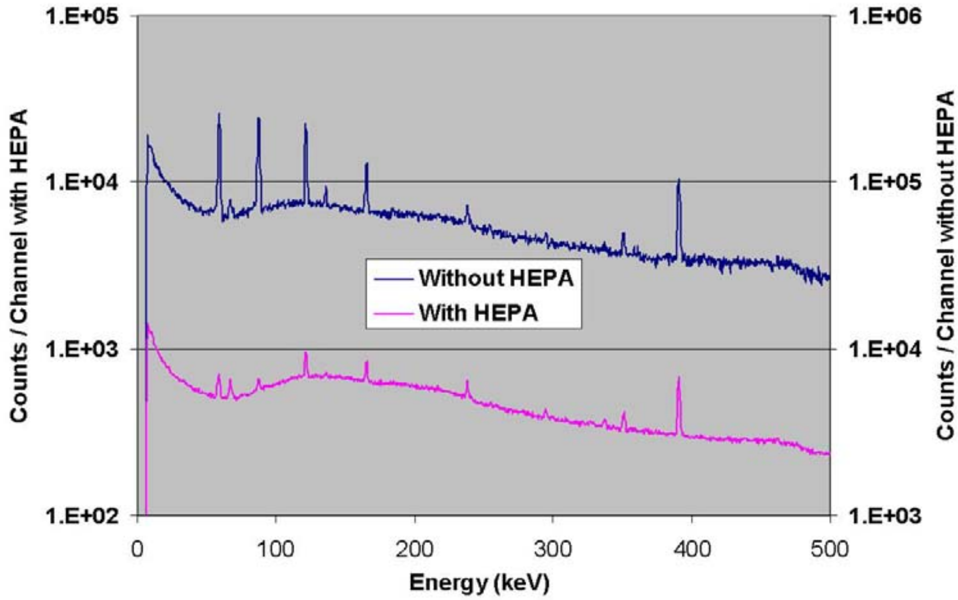

Figure 3. Spectra of a mixed-gamma-ray standard measured twelve inches from the face of the HPGe detector with and without the HEPA filter and its housing. Data are normalized to the same count time. Note that Counts / Channel data in are on different scales so the attenuation of low-energy gamma rays by stainless steel shell of the HEPA filter and its housing can be more clearly seen. Data taken with the HEPA and its housing are offset one order of magnitude lower.

After applying ISOTOPIC's attenuation corrections, the ratio of shielded to the unshielded results for these standards is near unity. For peaks bracketing Pu-238's 153 keV peak, the shielded to unshielded ratios were 1.108+-6.5% and 1.59+-8.1% at 122 and 166 keV peaks in the standard, respectively. For peaks bracketing Pu-239's 375 keV peak, peaks in the standard at 344 and 392 keV gave shielded to unshielded ratios of 0.922+-1.1% and 0.960+-4.9%, respectively. The counting statistics cited above are well within the estimated method uncertainty of 15%. These ratios confirm that the ISOTOPIC model provides appropriate attenuation corrections for holdup measurements of HEPA filters in the stainless steels used for the filter's shell and its glovebox housing.

Comparisons of results from spectra analyzed using ISOTOPIC data-analysis approach yielded results that were consistently within 2% of the MC&A value for a "known" Pu-239 source.

Separate calculations of geometry and attenuation corrections using MicroShield show agreement within a few percent to those given by ISOTOPIC for drums containing Pu-239.
The calculations determined

- geometry corrections (relative to a point source at 12 inches), and
- attenuation corrections for 55-gallon drums.

This independent comparison gave results that agreed within a few percent of those calculated by ISOTOPIC. The comparisons presented in Table 1 are for gamma-rays at 375 keV; this is one of the primary gamma-ray energies often used for nondestructive analysis of Pu-239.

Correction Factors	**MicroShield At 375 keV**	**Isotopic At 375 keV**
Air Gap Attenuation	1.011	1.012
Drum Wall Attenuation	1.150	1.132
Matrix Attenuation	1.213	1.217
Geometry	16.012	16.600
Combined C.F.	22.582	23.143

Table 1. A comparison of ISOTOPIC's geometry and attenuation correction factors to those given by MicroShield for Pu-239 dispersed in a 55-gallon drum.

GloveBox Process Holdup Measurements

The rooms that enclose the VE glovebox trains provide excellent contamination control, but they offer little space for the assay equipment, the portable HPGe detector on a detector positioning cart. Since the facility layout requires the detector to be too close to the glovebox to treat the measurements as "far-field," the glovebox to be assayed is divided into sections.

Figure 4. VE glovebox train at a Savannah River Site facility.

Detector collimators are set to give wide fields of view (FOV) that overlap at the predefined monitoring locations, and the results for each section are summed. Since the distribution of radioactive material is not known, the conservative assumption is made that all of the material is on the back wall of the glovebox rather than at locations closer to the detector. ISOTOPIC provides the geometry correction factor (a plane source at the distance to the back wall relative to the geometry of a point source at twelve inches) and an attenuation correction factor (glove box window).

RESULTS AND DISCUSSION

Conservatively high estimates of the radioactivity present in the gloveboxes are assured since 1) all of the activity is assumed to be at the furthest distance, and 2) results from overlapping views are summed. These results are multiplied by a factor of two for further conservatism. Figure 5 shows that this conservatism has not caused difficulties for facility operators. That is, the mass of Pu-239 reported following our assays has always been well below facility process holdup limits.

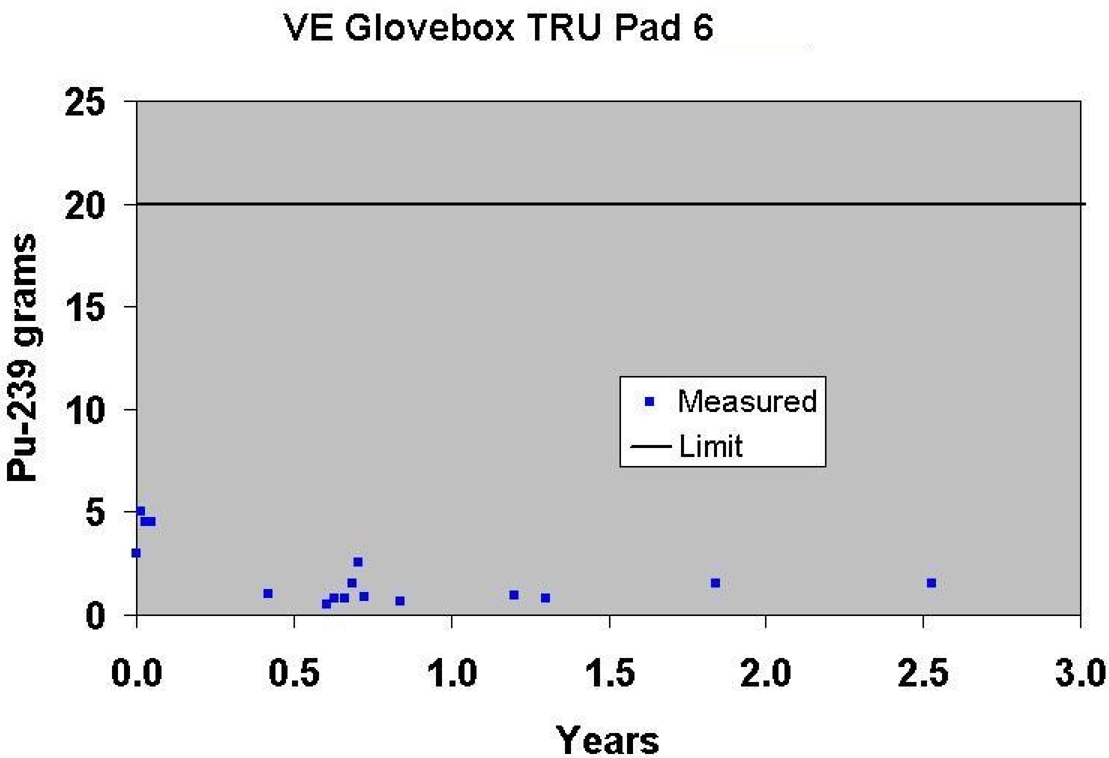

Figure 5. Measured holdup values for Pu-239 in a VE glovebox compared to facility operating limits.

Plutonium-239 is usually the nuclide of interest; however, Pu-241, Np-237 (including its daughter Pa-233) and Pu-238 (if detected) are typically assayed. Cs-137 and Co-60 may also be detected but are not reported since they do not contribute to the Pu-239 Fissile Gram Equivalent or Pu-239 Equivalent Curies. HEPA filters, drums and waste boxes are also assayed by the same methodology.

REFERENCES

1) American National Standard Calibration and Use of Germanium Spectrometers for the Measurement of Gamma-Ray Emission Rates of Radionuclides, ANSI N42.14-1999.

2) Genie2K Version 2.0 Canberra Industries

3) ISOTOPIC Version 2.0.6 Ametek/ ORTEC

4) Microshield Version 6.10, Framatome ANP, Inc.