

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

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U. S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied: 1. warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or 2. representation that such use or results of such use would not infringe privately owned rights; or 3. endorsement or recommendation of any specifically identified commercial product, process, or service. Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

May 14, 2007

ANALYSIS OF SPECIAL WASTE CONFIGURATIONS AT THE SRS WASTE MANAGEMENT FACILITIES

V.R. Casella and R. A. Dewberry
Savannah River National Laboratory
Aiken, SC 29808

ABSTRACT

Job Control Waste (JCW) at the Savannah River Site (SRS) Solid Waste Management Facilities (SWMF) may be disposed of in special containers, and the analysis of these containers requires developing specific analysis methodologies. A method has been developed for the routine assay of prohibited items (liquids, etc.) contained in a 30-gallon drum that is then placed into a 55-gallon drum. Method development consisted of system calibration with a NIST standard at various drum-to-detector distances, method verification with a liquid sample containing a known amount of Pu-238, and modeling the inner container using Ortec Isotopic software. Using this method for measurement of the known standard in the drum-in-drum configuration produced excellent agreement (within 15%) with the known value. Savannah River Site Solid Waste Management also requested analysis of waste contained in large black boxes (commonly 18'x12'x7') stored at the SWMF. These boxes are frequently stored in high background areas and background radiation must be considered for each analysis. A detection limit of less than 150 fissile-gram-equivalents (FGE) of TRU waste is required for the black-box analyses. There is usually excellent agreement for the measurements at different distances and measurement uncertainties of about 50% are obtained at distances of at least twenty feet from the box. This paper discusses the experimental setup, analysis and data evaluation for drum-in-drum and black box waste configurations at SRS.

INTRODUCTION

Job Control Waste (JCW) at the Savannah River Site (SRS) Solid Waste Management Facilities (SWMF) may be disposed of in special containers, and the analysis of these containers requires developing specific analysis methodologies. The JCW must be analyzed for TRU nuclides to determine if the waste is below the TRU limit (100 nCi/gram). An Ortec portable ISOCART System is used for the measurements using a high-purity germanium detector with a lead shield. Canberra Genie 2000 and Ortec Isotopic software are used for spectral analysis, drum calibration and attenuation corrections. A method has been developed for the routine assay of prohibited items (liquids, etc.) contained in a 30-gallon drum that is then placed into a 55-gallon drum. Method development consisted of system calibration with a NIST standard at various drum-to-detector distances, method verification with a liquid sample containing a known amount of Pu-238, and modeling the inner container using Ortec Isotopic software. Using this method for measurement of the known standard in the drum-in-drum configuration produced excellent agreement (within 15%) with the known value. Solid

May 14, 2007

Waste Management also requested analysis of waste contained in black boxes (commonly 18'x12'x7') stored at the SWMF. These boxes are frequently stored in high radiation areas; therefore, the background radiation from other black boxes and waste containers must be mitigated for each analysis. A detection limit of less than 150 fissile-gram-equivalents (FGE) of TRU waste is required for the black-box analyses. The analysis configuration was modeled and, if possible, at least two analyses are performed at different detector-to-box distances. The geometry correction is reduced as the detector-to-box distance is increased, while the counting statistics and background uncertainties are increased as this distance increases. There is usually excellent agreement for the measurements at different distances and measurement uncertainties of about 50% are obtained at distances of at least twenty feet from the box. This paper discusses the experimental setup, analysis and data evaluation for drum-in-drum and black box waste configurations at SRS.

MEASUREMENT PROCESS

Drum-in-Drum Assays

Figure 1 shows the side-view of the 55-gallon drum on the assay turntable and the top view of the 30-gallon drum inside the 55-gallon drum. The prohibited assay item is inside the 5-gallon container shown inside the 30-gal/55-gal drum-in-drum configuration.



Figure 1. Drum on counting turntable (left) and item in 30-gal/55-gal drum-in-drum configuration.

An Ortec portable ISOCART System was used for the measurements using a 40% germanium detector with a 0.625-in lead shield. Canberra Genie 2K and Ortec Isotopic software were used for spectral analysis, drum calibration and attenuation corrections. Energy calibration data were also collected. The collimated HPGe detector was positioned on the ISOCART (Figure 2) stand at a known calibration distance for drum-in-drum assays. The drum was rotated to ensure the assay was representative of the drum contents.



Figure 2. Isocart system used for the analyses.

The calibration of the detectors used for this method is described in technical report “Nondestructive Assay Efficiency Calibration of HPGe Detectors for the Ortec Isotopic Method”, WSRC-TR-2005-00431. The HPGe detector was calibrated using a NIST-traceable mixed-gamma-ray point source obtained from Analytix, Inc. with certified gamma-ray emission rates for the radionuclides in the standard with overall uncertainties of 2.6 to 3.3% in the energy range from 59 keV to 1836 keV. The point source was positioned in the inner drum where the prohibited item is placed and at various distances (4, 12 and 36 inches) from the face of the high purity germanium detector and this efficiency data was then input into ISOTOPIC. ISOTOPIC makes additional corrections to account for differences in the geometry of the prohibited item and container and in attenuation between the calibration point-source standard and the object being measured.

Results for a three milliliter Pu-238 solution that was analyzed at SRNL are given in Table 1. The solution was contained in a vial that was put inside the drum-in-drum configuration and then analyzed by the present method.

Table 1. Comparison of drum-in-drum analyses with known activity of Pu-238.

Analysis	Pu-238 (dpm)	Uncertainty (2σ)	% Relative Difference
SRNL	2.53E+10	0.078E+10	
Drum-in-drum (12")	2.16E+10	0.11E+10	15
Drum-in-drum (36")	2.35E+10	0.12E+10	7.1

As shown in Table 1, the drum-in-drum results for the solution agreed with the known value to within 15% at a detector distance of twelve inches and to within 7.1% at a detector distance of thirty-six inches. As the detector-to-drum distance increases, the measurement of the exact distance is less critical and the method is expected to be more accurate as seen by the results presented.

ISOTOPIC provides results for selected gamma-emitting nuclides. For each analysis the inner container that contains the prohibited item (within the 55-gallon and 30-gallon

May 14, 2007

drums) must be configured and pertinent parameters (weight, dimensions, etc) must be entered into the software as shown in Figure 3.

Figure 3. Isotopic prohibited item container configuration.

Thickness 0.060 in. Measurement Series Sample title.
 Sample ID:
 Container height 14.7 in.
 Height 7.4 in. Diameter 10.2 in. Standoff 10.2 in.
 Cylinder was rotated when counted
 OK
 Print form
 Cancel
 Reset Config.

Container	Detector position	Collimator
Diameter 10.2 in.	Detector height 7.4 in.	Select
Height 14.7 in.	Standoff 10.2 in.	ID 5-8ths-Pb
Material Steel	Matrix <input checked="" type="radio"/> Gross Weight <input checked="" type="radio"/> Density	Material: Lead
Density 7.8 g/cc	Material Combustible	Collimation: 1.5 in.
Thickness 0.060 in.	Density 0.300 g/cc	Thickness: 0.6 in.
Tare wt 0.0 lb	Wt fraction U 0.001	Diameter (ID): 4.3 in.

Results for the nuclides-of-interest are obtained and the nuclides that are not measured can be scaled based on concentrations for a known waste stream. If a TRU nuclide is not detected for the count time used (usually one hour), the minimum detectable activity (MDA) is used for the determination. In practice, nearly all of the drum-in-drum assays are done at a distance of thirty-six inches, and the method accuracy also includes counting statistics and attenuation uncertainties that are determined by the ISOTOPIC software. Reported uncertainties are usually better than $\pm 20\%$.

Black Box Assays

SRNL was requested to provide an estimate of the plutonium content of nine black boxes stored at the TRU Pad 7-13 Area of the Waste Management Facilities to verify the plutonium content. Two of the boxes were in a high radiation area.

In this case, an Ortec portable ISOCART System was used for the measurements using a 20% germanium detector with a 0.50-in lead shield. Canberra Genie 2K and Ortec Isotopic software were used for spectral analysis, box calibration and attenuation corrections. The collimated HPGe detector was positioned on the ISOCART stand at distances from four feet to thirty feet from the box, and each box was counted such that the counting statistics uncertainties were less than 5% or a minimum detectable activity (MDA) was used.

The ISOTOPIC configuration for the black box measurements (18' x 12' x 7' box) is presented in Figure 4.

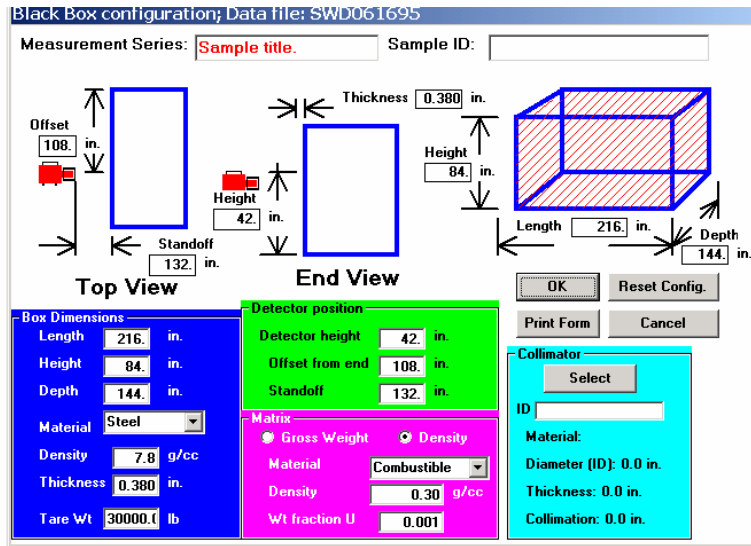
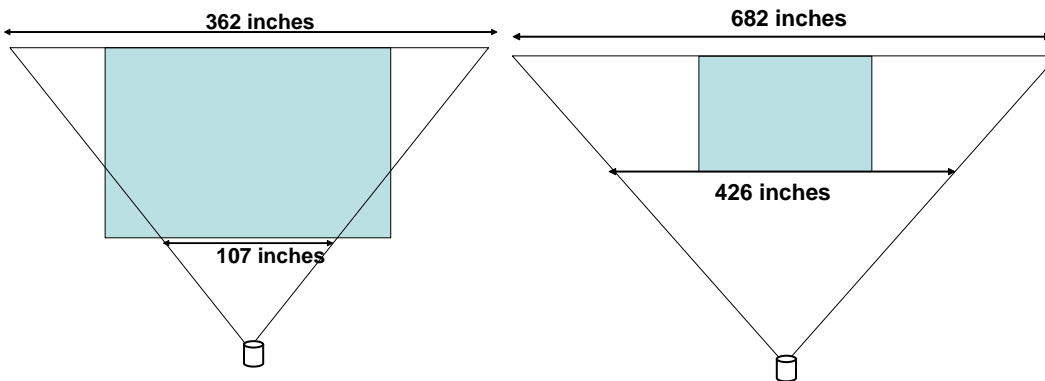


Figure 4. Isotopic black box container configuration.

The detector-to-box distances depended on the size of the box and the position of the box relative to other boxes in the immediate vicinity. It was advisable to have the detector as far away from the box as possible to reduce the geometry correction uncertainty as long as the counting statistics uncertainties and background radiation were acceptable. If possible, the detector positioning was done so that other boxes that may contain TRU nuclides of interest were not in the detector field-of-view. The box sizes varied from 52" long x 50" wide x 60" high to 216" long x 144" wide by 84" high; weights varied from 6,000 lbs to 44,000 lbs. As shown is the fields-of-view (FOV) in Figure 5, all of the 18' x 12' x 7' box is counted for a detector-to-box distance of 20 feet, while about 90% of the box is counted for a detector-to-box distance of 5 feet. For the detector-to-box distance of 5 feet, the box is counted on both sides so that the entire box is assayed.



May 14, 2007

Figure 5. Detector fields-of-view for detector-to-box distances of 5 feet (left) and 20 feet (right) for a 18' x 12' x 7' black box.

The 413-keV and 153-keV gamma rays were used for Pu-239 and Pu-238 assays, respectively. Since the 153-keV gamma ray is lower energy, "infinite thickness" was usually reached; therefore, some of the activity was estimated by the Ortec Isotopic

Software based on the assumption that the box contents were distributed uniformly throughout the box.

The count times were usually from 30 minutes to four hours. However, for a 16-hour count in a low-background area, a detection limit for Pu-239 of 6 grams was achieved. The maximum gross weights for the boxes were used for attenuation corrections, rather than an average weight for these types of boxes and waste streams. This makes these assays very conservative (an increase of about 50%). Also, twice the uncertainty of the measurement was added to the measured value to add additional conservatism.

Several assumptions were made for these measurements:

- The gamma-ray attenuation was calculated for a steel box thickness of 0.375 inches, and the material in the box (steel, plywood, etc) was assumed to be distributed uniformly throughout the box;
- All of the measured activity was assumed to be only from the box.
- There were no extremely high density items in the box that would essentially prevent the gamma rays from being detected. Very high density items would result in underestimating the plutonium present.

This method produced a very conservative estimate of the plutonium present in the large black box containers to show that the fissile-gram-equivalents (FGE) of TRU waste present was less than the 150 gram limit.

CONCLUSIONS

Specific analysis methods were developed for the analysis of TRU waste prohibited items in the drum-in-drum (30-gallon drum inside 55-gallon drum) configuration and for TRU waste in large black boxes. Providing that counting statistics uncertainties are less than 5% and the job-control-waste does not contain high-density items, the drum-in-drum method uncertainty is less than 15% and the black-box method provides a very conservative estimate of the TRU waste present to show that the fissile-gram-equivalents (FGE) of TRU waste present is less than the 150 gram limit.

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. ISOTOPIC Version 2.0.6 Ametek/ ORTEC
3. Genie2K Version 2.0 Canberra Industries
4. R.A. Sigg, V. R. Casella and R. A. Dewberry, WSRC-TR-2005-0431, "Nondestructive Assay Efficiency Calibration of HPGe Detectors for the Ortec Isotopic Method."