PU-238 ASSAY PERFORMANCE WITH THE CANBERRA IQ3 SYSTEM

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ABSTRACT

Canberra Industries has recently completed a demonstration project at the Westinghouse Savannah River Site (WSRC) to characterize 55-gallon drums containing Pu-238 contaminated waste. The goal of this project was to detect and quantify Pu-238 waste to detection limits of less than 50 nCi/g using gamma assay techniques. This would permit reclassification of these drums from transuranic (TRU) waste to low-level waste (LLW). The instrument used for this assay was a Canberra IQ3 high sensitivity gamma assay system, mounted in a trailer. The results of the measurements demonstrate achievement of detection levels as low as 1 nCi/g for low density waste drums, and good correlation with known concentrations in several test drums. In addition, the data demonstrates significant advantages for using large area low-energy germanium detectors for achieving the lowest possible MDAs for gamma rays in the 80-250 keV range.

Prepared for the 5th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference; Jan 14-16, 1997; Salt Lake City, UT.

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INTRODUCTION

The goal of the mobile waste characterization service provided by Canberra was to demonstrate the capabilities of the assay system to meet WSRC requirements for characterizing "Suspect TRU" category waste. The IQ3 system contains three 45% relative efficient HPGe detectors and three large area Low Energy Germanium (LeGe) detectors for high sensitivity counting. Drums are loaded onto an automated conveyor system, moved to the counting chamber, loaded with a fork lift mechanism mounted on the shield door and placed on a turntable in a six inch thick steel shield counting chamber. Following completion of the count, the drum is unloaded onto the exit side of the conveyor system. All operations may be controlled from the operator's console in the control room. Results are computed and printed or displayed within two minutes of count completion.

The IQ3 was placed on a waste storage pad in the solid waste management area of the WSRC site. The pad is covered with a pole tent structure and is used for TRU waste drum storage. The pad is considered a Radiological Buffer Area (RBA) requiring both security and radiological controls. Canberra was required to provide all utilities and trailer services, while WSRC provided drum handling and safety support.

Two sets of drums were selected for counting, a group of unknown process drums, and a set of known test drums. The unknown drums were selected from low activity, low density waste drums stored on site, and consisted of six drums marked "Pad Storage" and 14 drums designated as "Culvert Storage". The Pad Storage drums were known to have originated from process facilities, but were believed to contain very low level (or possibly no) activity. The Culvert Storage drums were so designated because of the possibility of containing more than 0.5 Ci TRU waste. All unknown drums had no detectable external radiation levels and weighed from 50 to 200 pounds. Drums were known to contain low density, low activity compactable waste such as protective clothing, plastic sheeting, breathing air line hosing, small hand tools and polyethylene bottles. The known test drums were prepared by WSRC personnel and contained known (to WSRC) amounts of Pu-238 in similar drum matrices.

All drums were counted on the IQ3 system for count times ranging from 15 minutes to one hour. Following the initial counting of all drums, system parameters were adjusted to account for WSRC specific conditions, and a second round of counting for selected drums was performed. Results are presented and discussed in the following sections.

Figure 1: Picture of IQ3 System

SYSTEM OPERATION

The IQ3 system contains three 45% relative efficient HPGe coaxial detectors for wide energy range detection and three 2800 sq. mm area LeGe detectors for low energy gamma/x-ray detection. The system is able to correct for non-homogeneous matrix distributions by employing photon transmission corrections, and identifies non-homogeneous source distributions by performing multi-spectral scaling as the drum rotates. The LeGe detectors can be used for Multi Group Analysis(MGA) when Pu or U isotopes are present. The system has a scale for weighing drums and calculates densities based on these weights. The data are analyzed with the standard Canberra Genie Waste Assay (GWAS) software.

The HPGe coaxial detectors are energy calibrated to about 0.5 keV per channel for a 4096 channel spectrum. Two drum analyses may be performed with these detectors: a sum of spectra analysis and an average segment analysis. The sum of spectra analysis sums the three detector spectra and analyzes the summed spectra for nuclide activity. This analysis assumes a uniform activity and matrix distribution throughout the drum and uses an efficiency which is interpolated from a set of calibration curves obtained from uniformly distributed activity in different density drums.

The average segment analysis routine uses the individual coaxial detectors and corrects each for matrix transmission. The transmission sources (Ba-133 is used to evaluate transmission in the 80 to 400 keV region) located opposite each coaxial detector are used to evaluate the matrix density in the region directly in front of the detector. Transmission peak intensities are compared to an empty drum transmission to develop correction factors for the matrix, using attenuation coefficients for lucite as the default values. The transmission corrected activity values for each of the three detectors are averaged to arrive at the total drum average segment activity.

The LeGe detectors are normally used to perform MGA on spectra with multiple Pu or U isotopes, and requires Pu-239 and Pu-241 to be present for Pu determinations. Since the demonstration project involved the measurement of Pu-238 only, MGA could not be used. As an alternative, the LeGe detectors were calibrated and configured to perform a sum of spectra analysis, as with the coaxial detectors, and were used as confirmation of HPGe coaxial detector results. The initial energy range for the LeGes was 0 to 300 keV, which was deemed optimum for Pu-238 measurements. Additional studies were performed to evaluate the LeGes when configured for use with transmission sources and for energies up to about 500 keV.

The GWAS software provides methods for assessing both source and matrix non-uniformity. The results of these analyses can be used to determine which analysis routine (sum of spectra or average segment) is optimum and to assign appropriate uncertainties due to non-uniformity. Multi-Spectral Scaling is used to determine radial source and/or matrix non-uniformities. For each detector, sample and transmission source counts are collected in eight radial segments as the drum rotates. For sample counts, a high count rate in one segment indicates a concentration of activity in that segment. Likewise, a low transmission source count in one segment indicates a high matrix attenuation and a possible non-uniform matrix distribution.

Nuclide results and transmission corrections, per detector, may be used to infer vertical non-uniformity for both source and matrix. High activity results for one detector indicates a concentration of activity in a vertical segment of the drum, and a low transmission factor for a detector indicates high absorption, and probable matrix concentration or variation of matrix material, in a vertical segment.

SYSTEM CALIBRATION

The IQ3 analysis system was calibrated to measure radionuclide activities and/or concentrations in 55 gallon drums. Efficiency calibrations are made with uniform source and matrix distributions. A uniform source distribution is simulated with line sources placed vertically in the drum in the center of a series of equal volume cylinders such that a uniform distribution is approximated when the drum is rotated. The line sources consist of mixed nuclides with photopeaks at intervals sufficient to calibrate over the energy range of interest. Four drums with various density materials are used to obtain efficiency vs density

calibrations. Calibration drums were standard DOT-17C containers, without liners. Due to license restrictions, Canberra is unable to possess sufficient Pu or U sources to perform calibrations or verifications with actual TRU materials, and must rely on customers to provide these verifications when possible.

DRUM COUNTING PROCEDURES

All drums were loaded on the conveyor and counted on the IQ3 system using the automated GWAS procedures. Count times were varied to obtain a range of detection limits. For the first round of counting, factory specified default parameters were used for data acquisition and analysis. Following review of this data, several modifications were made to address WSRC specific conditions. For example, initial Pu-238 quantification was accomplished using the 152 keV line only, since other lines were considered as likely to be interfered with by other nuclides or had abundances too low to be of use. Subsequent analyses also used the Pu-238 line at 99.9 keV, since it was determined that interfering nuclides were absent from the sample and test drums. Efficiencies were adjusted to account for WSRC drum liners, which were absent from Canberra calibration drums. The average segment analysis routine was selected for drum analyses, based on indications of non-uniformity of materials in drums. Correct tare weights for the WSRC drums were entered as required. Following these modifications, a second round of counting of selected drums was performed to establish final accuracy, precision and sensitivity values for the IQ3.

DRUM COUNTING RESULTS

Results of measurements of unknown drums are shown in Table 1. All six drums labeled as Pad Storage drums showed no detectable Pu-238 activity. The MDA levels ranged from 1.7 to 4.9 nCi/g Pu-238. The Culvert Storage drums contained from non-detectable to 2000 nCi/g Pu-238, but all had less than 0.5 Ci total activity, the requirement for Culvert Storage designation. In addition to Pu-238, several drums contained very low levels of Cs-137, K-40 and/or Ra-226 daughters Pb-214 and Bi-214 (these concentrations were on the order of a few pCi/g). These results confirmed that WSRC had been conservative when classifying and storing "Suspect TRU" waste, and that most of this category of drums could be designated as low level waste and dispositioned accordingly.

Table 1 Summary of TRU waste drum measurements on unknown drums.

Pad Storage Drums	Pu-238 nCi/g	Culvert Storage Drums	Pu-238 nCi/g
557460	<4.6	608969	<8.3
557459	<4.1	608967	930 +/- 120
557475	<4.9	608970	1180 +/- 190
557521	<1.7	608971	<5.9
557439	<3.2	608974	1800 +/- 220
557454	<2.8	608979	220 +/-35
		608978	1950 +/- 260
		608975	80 +/- 12
		608968	<6.7
		608976	<2
		608973	1200 +/- 160
		608965	<5.3
		608977	430 +/- 80
		608972	510 +/- 65

All values are Pu-238 nCi/g concentrations. The error is a 2 sigma counting error

Table 2 shows the results of final accuracy, precision and sensitivity determinations using the WSRC prepared test drums. The values in Table 2 are those obtained following the initial round of counting and subsequent system realignment for WSRC specific conditions. Due to project time limitations, Canberra was unable to perform the complete set of replicate counts required by the TRU Quality Assurance Program Plan; however, the data collected indicates that the IQ3 easily meets the Quality Assurance Objectives(QAOs) for these drums. These results show that the system is accurate to better than +/- 10% for concentrations ranging from 50 to 200 nCi/g Pu-238. The precision (reproducibility) at 50 nCi/g was good, with a standard deviation of +/- 4.1 % and all measurements within the S.D. Detection limits on the order of a few nCi/g are achievable in a 30 minute

Table 2a. Summary of TRU test drum measurements for accuracy.				
Test Drum #	**CI Result Activity (mCi)**	**Actual Activity (mCi)**	**CI Result Concen (nCi/g)**	**Actual Concen (nCi/g)**
1	2.08 +/- 0.34	2.07	100 +/- 16	97.4
1	1.99 +/- 0.33	2.07	98 +/- 16	97.4
2	0.89 +/- 0.17	1.00	46 +/- 9	50.1
2	0.94 +/- 0.18	1.00	48 +/- 9	50.1
2	0.95 +/- 0.18	1.00	49 +/- 9	50.1
2	0.91 +/- 0.17	1.00	46 +/- 9	50.1
2	0.92 +/- 0.18	1.00	47 +/- 9	50.1
3	3.23 +/- 0.50	3.13	208 +/- 32	194.4
3	3.31 +/- 0.52	3.13	213 +/- 34	194.4
3	2.95 +/- 0.38	3.13	190 +/- 35	194.4
3b	1.41 +/- 0.23	1.30	41 +/- 7	37.4
3b	1.33 +/- 0.24	1.30	38 +/- 7	37.4
3b	1.38 +/- 0.23	1.30	40 +/- 7	37.4

The table shows measured vs actual activities for the three test drums. Errors are 2 sigma counting errors only. Drum 3b was prepared following CI reconfiguration and recounting of the original test drums.

Table 2b. Summary of TRU test drum measurements for precision.
Drum 2 Activity (mCi)
0.918
0.905
0.950
0.889
0.936
Mean = 0.919 S.D. = 0.038

Drum 2 counted five times for 20 min. each for reproducibility. All measurements fall within one S.D. of the mean.

Table 2c. Summary of TRU test drum measurements for sensitivity.		
Count Time (min)	**MDA value Activity (mCi)**	**MDA Value Concen (nCi/g)**
10	<0.161	<10.1
20	<0.112	<7.20
30	<0.088	<5.58
60	<0.062	<3.86

Drum 608965 recounted to obtain an a priori estimate of system sensitivity for different count times. Drum density is 0.08 g/cc.

count with the coaxial detectors. Counting uncertainties for these measurements are on the order of +/- 20% (95% confidence level) for a 30 minute count. Other uncertainties have been estimated at +/- 30%, based on review of all count parameters and estimated systematic uncertainties. Total propagated uncertainties are estimated to be in the +/- 50% range for these drums.

Note: The results for the initial round of counting showed that the system was biased low by -8% to -24% for the test drums, values which are still within the acceptable range for the QAO for accuracy. Realignment of the system resulted in increasing the response of the system by about 15%.

CONCLUSIONS

The results of the TRU waste characterization demonstration project verifies that the Canberra IQ3 system will measure Pu-238 activity in low activity, low density waste drums with high accuracy, precision and sensitivity. The system allows for segregation of TRU waste from low level waste for Pu-238 contamination and meets the QAOs for accuracy, precision, sensitivity and uncertainty for TRU waste characterization of these drums. Based on counting results, Canberra estimates the dynamic range of the standard IQ3 for counting Pu-238 to be from microgram quantities to approximately 1 gram Pu-238.