Final Evaluation of Characterizing Pipe-Over-Pack Containers Using High Efficiency Neutron Counters - 9086

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ABSTRACT

Nondestructive assay (NDA) measurements of Transuranic (TRU) waste at Los Alamos National Laboratory (LANL) packed in Pipe-Over-Pack Containers (POC) contain a number of complexities. The POC is highly attenuating to both gamma rays and neutrons which presents a difficult waste matrix for correct quantification of material in the container. Currently there are a number of POC containers at LANL that require evaluation for shipment to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM. Updated data has been evaluated that finalizes the evaluation of characterizing Pipe-Over-Pack Containers.

Currently at LANL, a single instrument has been used to explore the appropriateness of both passive neutron and quantitative gamma ray methods for measuring POC's. The passive neutron approach uses the Reals coincidence count rate to establish plutonium mass and other parameters of interest for TRU waste. The quantitative gamma ray method assumes a homogeneous distribution of radioactive source material with the surrounding material throughout the drum volume. Drums are assayed with a calibration based on the known density of the matrix. Both methods are supplemented by a simultaneous isotopic measurement using Multi-Group Analysis (MGA) to determine the plutonium isotopic composition. If MGA fails to provide a viable isotopic result Fixed Energy Response function Analysis with Multiple efficiencies (FRAM) has been used to replace the MGA results. Acceptable Knowledge (AK) may also be used in certain instances.

This report will discuss the two methods in detail. Included in the discussion will be descriptions of the setup parameters and calibration techniques for the instrument. A number of test measurements have been performed to compare HENC data with certified historical data. Empty POCs loaded with known sources have also been measured to determine the viability of the technique. A comparison between calorimetry data, historical measurements and HENC data will also be performed. The conclusion will show that the current calibration on the HENC units is viable for analysis of POCs.

INTRODUCTION

Nondestructive assay techniques measure radiation emitted from nuclear materials without altering their physical or chemical state. At Los Alamos National Laboratory (LANL) there are a number of Pipe OverPack containers containing large amounts of nuclear material. It is an important part of the Transuranic Waste Program at LANL to quickly remove these high activity waste drums content material from the site.

Measuring transuranic waste placed in POCs poses a number of technical issues for NDA. The Pu contaminated waste cans are first placed into a stainless steel pipe with a thickness of 0.25 inches. This pipe is centered in a 55 gallon drum that is then surrounded with fiberboard/plywood dunnage. The stainless steel pipe and the packaging material cause significant attenuation for gamma sprectroscopy measurements. Neutron measurements are not significantly affected by the stainless steel but the dunnage contains a high amount of hydrogenous material.

The purpose of this report is to describe the effectiveness of the methods used to measure POCs at LANL in detail. Included in the discussion will be descriptions of the POC, instrument setup parameters, calibration techniques and analysis restrictions for the methods. An evaluation of waste measurements containing plutonium and source measurements contained in POC drums will be presented in order to separate container/dunnage and matrix effects on the measurements. In the context of this report, matrix effects refer to the material associated with the chemical processing of the plutonium contained in the waste.

PIPE OVERPACK

The standard pipe overpack identified in CH-TRAMPAC Revision 2 was used for the test measurements. There are two types of POCs identified in the TRAMPAC, a 6 inch pipe component and a 12 inch pipe component. For this evaluation the material was placed in the 12 inch pipe component. Figures 1 and 2 below show the actual configuration of the POC.

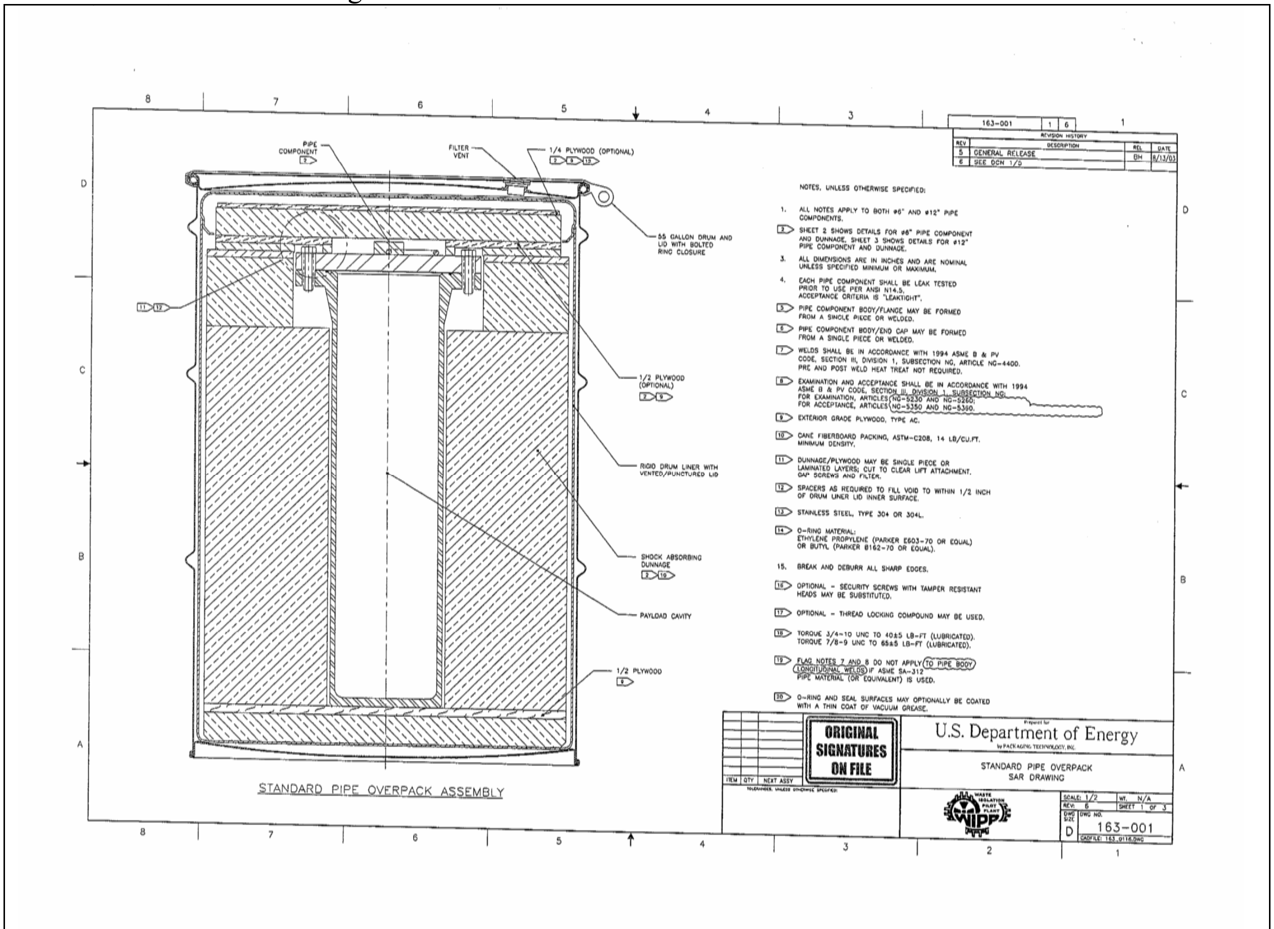

Figure 1: Standard pipe overpack assembly as shown in the CH-Trampac Rev. 2 [1]. All notes in the diagram apply to the 6 inch and 12 inch pipe overpack containers.

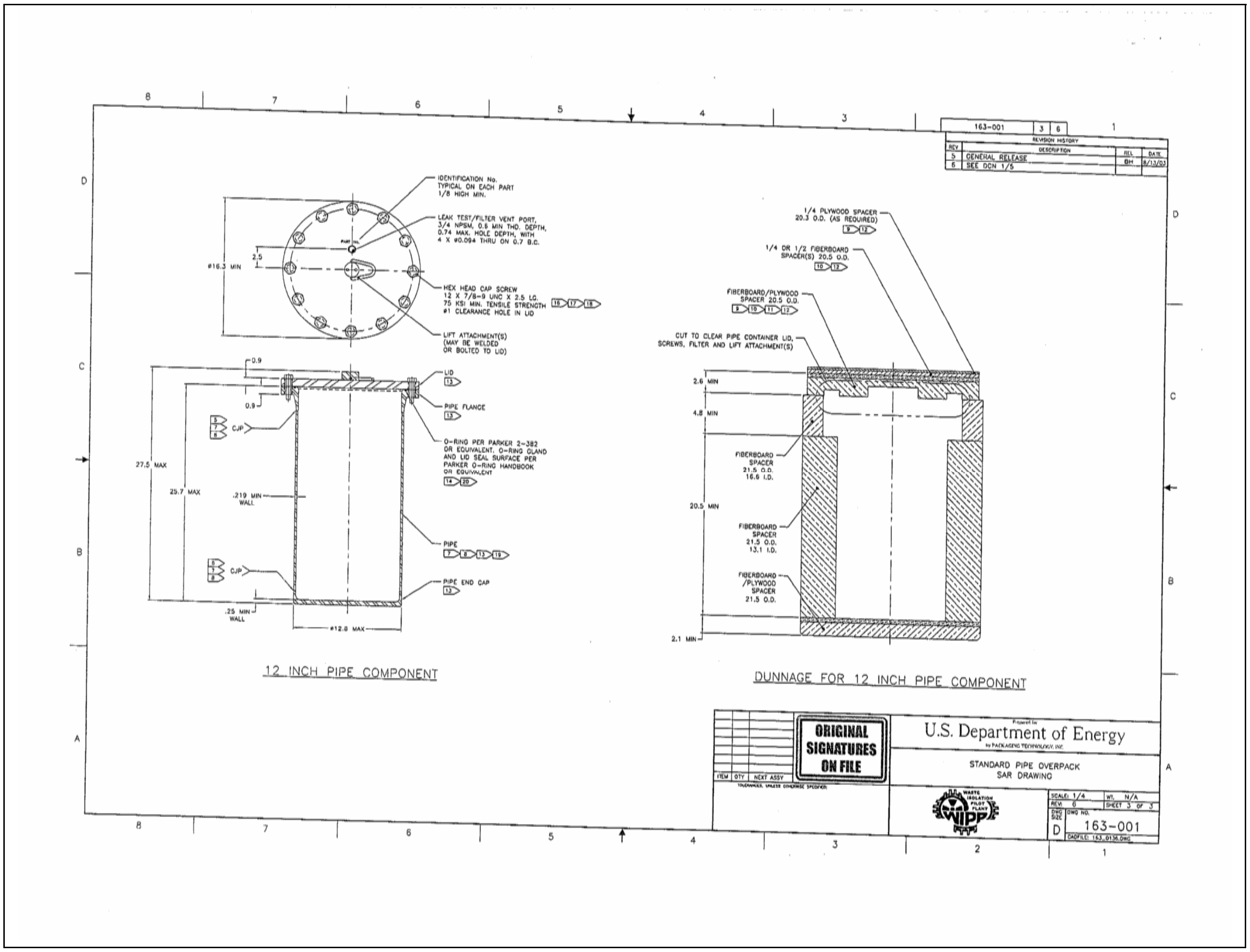

Figure 2: 12 inch pipe component from the CH-TRAMPAC Rev. 2 [1].

The waste placed in POC containers was produced at the Los Alamos National Laboratory. For a complete evaluation of the technique different types of waste streams were tested. One of the waste streams consisted of mainly Pu-238 by mass. Another stream consisted of primarily Pu-239 by mass with some of the waste containing a large amount of Am-241. Other radionuclides can also be present in the material in significant amounts such as Np-237, but the main contributors to the activity will be either Pu-238, Am-241 or Pu-239.

NDA METHODS USED TO MEASURE PIPE OVERPACK CONTAINERS

The LANL HENC (HENC #2), shown in Figure 3, is a hybrid NDA instrument based on the High Efficiency Neutron Counter (HENC) with an integrated gamma system. The HENC was not originally designed to house a gamma-ray spectroscopy system. The design was modified in order to improve the capability of the instrument. With the added capability the system integrates the two NDA counting modes: passive neutron and gamma-ray assay.

Figure 3: High Efficiency Neutron Counter at Los Alamos National Laboratory (HENC#2).

High Efficiency, Add-A-Source, Passive Neutron Coincidence Counting Modality

The HENC #2 passive neutron counter employs neutron coincidence and multiplicity counting of 208-liter drums. The counter utilizes 113 He-3 proportional detectors, divided into 16 detector banks, arranged in a 4-π geometry about the assay cavity. The nominal assay cavity is 81 cm wide by 86 cm long by 102 cm tall. An Add-A-Source (AAS) Matrix Correction assembly has been incorporated into the counter for neutron moderation and absorption correction.

The neutron portion of the instrument is calibrated [2] using NIST-traceable standards to create a calibration curve that compares the coincidence count rate to the Pu-240 effective mass. The Pu-240 effective mass is related to the spontaneously fissioning isotopes of plutonium by the following equation

$$m_{Pu240Eff} = 2.52m_{Pu238} + 1.0m_{Pu240} + 1.68m_{Pu242} \quad \text{(Eq. 1)}$$

The calibration curve is used to calculate a Pu-240 effective mass value for neutron measurements of waste drums. These values, in turn, are combined with isotopic measurements and AAS corrections to determine the total plutonium mass. The neutron calibration was performed using standard 55 gallon drums. Pipe over-pack containers were not used for the neutron mass calibration.

Gamma-Ray Assay Modality

The gamma portion of the HENC system uses a Broad Energy Germanium (BEGe) gamma-ray detector. This detector is mounted in one of the sidewalls of the counter perpendicular to, and pointing towards, the vertical axis of the sample drum in the counter cavity. The detector is mounted such that it can be withdrawn from the counter side wall when required. The detector is firmly positioned in the sidewall upon reassembly for routine gamma ray assays.

The spectrum from the BEGe gamma detector is processed by the acquisition electronics that are controlled by an external system computer. Both the passive neutron and the gamma-ray signals are processed and analyzed by the Canberra NDA 2000 waste assay software package.

The gamma calibration [3] method requires both an energy calibration and an efficiency calibration. The energy calibration is straightforward and requires that the unique peaks associated with known radionuclides fall within a specified energy range. The efficiency calibration requires measurement of gamma ray count rates from isotopes of known activity at energies between 60 keV and 1200 keV as a function of the density of the matrix material. Five matrix drums were used to establish the efficiency calibration: foam, homasote, particle board, sand, and concrete. The densities of the calibration drums range from 0.002 g/cc to 2.1 g/cc. Am-241/Eu-152 line sources that extend the full axial length of the 208-liter drums are used for the calibration. The sources are placed in a configuration such that, when the drums are rotated during the calibration measurements, the sources resemble a uniform source distribution. With this information, measurement of an unknown source in similar matrices yields the activity of each isotope. The gamma calibration was performed using standard 55 gallon drums. Pipe overpack containers were not used for the gamma calibration. As stated earlier, the underlying assumption of the gamma-ray assay modality is that matrix, dunnage, and source materials are uniformly distributed throughout the drum volume. Since POCs consist of small cans with isolated Pu chunks dispersed non uniformly in a processing matrix and separated by a 0.25' pipe from the dunnage, this assumption is seriously violated. However, the modality was tested here in order to determine whether aspects of the gamma method could be used to assay POC drums.

Additionally, the BEGe gamma-ray detector is used to measure the plutonium isotopic composition of the waste during each drum assay. Determination of the plutonium and Am-241 isotopic distribution is performed with Multi Group Analysis software under the control of NDA 2000. This measurement is performed concurrently with the quantitative gamma ray measurement. FRAM isotopic analysis, when used, was applied during later reanalysis.

POC EVALUATION DESCRIPTION

The POC evaluation consisted mainly of three different comparisons. The first was comparing historical measurements of actual POC waste with HENC results. The second portion of the evaluation was to load the central pin in a POC container with standards of known plutonium mass and to determine the accuracy and precision of the measurements. The third portion of the evaluation compared the HENC results with current calorimetry results. Accuracy estimates for the HENC measured values are reported as Percent Recovery (%R) using the historical baseline values and standard values as expressed as baselines. Precision estimates are reported as Percent Relative Standard Deviations (%RSD).

The POC waste evaluation consisted of 4 different containers. The first container was a low Pu mass drum with approximately 2.6 grams of MT-52 waste. MT-52 waste is defined as material containing 4.0 to 7.0 % Pu-240. The second container was a mid-range Pu mass drum with approximately 47.2 grams of MT-52 waste. The third container was a high mass drum containing approximately 111 grams of MT-52 waste. The last container was a high activity mass drum and contained approximately 5.1 grams of MT-83 (Heat Source) waste. MT-83 waste is defined as material containing 83% to 89% Pu-238. These mass values were either determined by neutron or gamma-ray NDA instruments. However the specific method of establishing the baseline values could not be confirmed. AK information was limited on the waste stream.

A total of eight measurements were made for each drum. Six of the assays were performed at the standard count time of 1800 seconds for the HENC. Two of the assays were set to count to precision with a maximum count time of 3600 seconds. A direct comparison of Total Pu mass was performed for the neutron measurements. A direct comparison of either Pu-238 or Pu-239 was performed for the gamma measurements.

The POC source evaluation was performed using one 12" POC container loaded with dunnage material and Pu sources of known activity placed in the center pipe. The sources consisted of PuO_2 uniformly dispersed in diatomaceous earth and contained in doubly encapsulated 1.95" diameter x 9.00" long stainless steel cans. Test measurements were performed by loading sources of known plutonium activity and composition into the POC drum with pipe and fiberboard dunnage but no other radioactive material present. Table 3 describes the standard masses used for each test run in the empty POC container. The neutron measurements were evaluated by a direct comparison of the Pu-240 effective mass value. The gamma results were evaluated by direct comparison of the Pu-239 or Pu-238 result.

A total of fourteen measurements were performed for the standard comparison. Six of the assays were performed at the standard count time of 1800 seconds for the HENC. Six of the assays were set to count to precision with a maximum count time of 3600 seconds. Two assays were also performed with a two hour count time. The difference between the HENC measured values the historical baseline values and the standard values are expressed as % Recovery (%R).

A total of 5 different drums were analyzed as part of the calorimetrly based comparison. Two of the drums contained MT-52 waste with one having 92.2 grams of total plutonium and the other having 117.5 grams as determined by calorimetry. The other three drums contained MT-83 with the following total plutonium masses 5.53 grams, 12.38 grams and 19.6 grams again determined by calorimetry. Gamma-ray isotopic measurements were also performed for each item. The neutron measurements were evaluated by a direct comparison of the Pu-240 effective mass value. The gamma results were evaluated by direct comparison of the Pu-239 or Pu-238 result.

POC WASTE EVALUATION RESULTS

The first tests performed on the HENC were the waste POC containers from historical data. The data obtained from the HENC assays was compared to historical data. Table 1 describes the neutron results for the waste POC container and table 2 describes the results for the gamma modality. In the Table, % Recovery is denoted as the ratio of the HENC results to the historical baseline values expressed as a percentage. The neutron results were based on standard neutron coincidence counting techniques. The results presented are compared to the WIPP/WAC requirements for calibration confirmation. These tests do not meet the full definition for a calibration confirmation as described by the WIPP/WAC but give a basis to evaluate the results.

Table 1: Neutron results from POC historical waste measurements. The values stated in this table are the average of six standard 1800 second measurements.

Waste Drum	Average Pu Mass Neutron	% Recovery	STDEV	%RSD
2.6 Gram MT-52*	3.9	151.3%	0.5	13.4
47.2 Gram MT-52	95.0	201.2%	12.1	12.7
111 Gram MT-52	145.8	131.4%	6.9	4.7
5.1 Gram MT-83	8.6	168.3%	1.7	2.0

* The 2.6 Gram drum designates that the mean value is from only five measurements.

The neutron results showed a large bias for the waste POC drums. The lowest average percent recovery value was for the 111 gram MT-52 drum. The percent relative standard deviation %RSD values would meet WIPP precision requirements. The upper limit for the %RSD value quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 (DOE/WIPP-02-03122 [2]) for six measurements is 14%. However the recovery values do not meet the performance requirements stated in the WAC. The neutron results were based on standard neutron coincidence counting techniques. The limits quoted from the WAC are intended for standard measurements but are applied to evaluate the instruments response.

Table 2: Gamma results from POC historical waste measurements. The values stated in this table are the average of six standard 1800 second measurements. The asterisk for the 2.6 gram drum designates that the mean value is from only five measurements.

Waste Drum	Average Pu Mass Gamma	% Recovery	STDEV	%RSD
2.6 Gram MT-52*	4.3	164.9%	0.4	9.0
47.2 Gram MT-52	82.7	175.1%	4.1	5.0
111 Gram MT-52	145.2	130.8%	3.7	2.5
5.1 Gram MT-83	5.6	110.0%	0.3	4.8

The gamma results showed a large bias for the waste POC drums. The lowest average percent recovery value was for the 5.1 gram MT-83 drum. The %RSD values for the gamma measurements would meet the WIPP waste acceptance criteria. However the recovery values do not meet the stated WAC requirements for three of the four drums. The MT-83 drum does meet the WAC requirements for %recovery and %RSD. Self-absorption correction techniques were applied for the gamma results on the POC waste drums.

POC STANDARD EVALUATION RESULTS

An empty POC container was loaded with known standards of known activity to evaluate the response of HENC#2. The standard mass loadings are described in Table 3.

Table 3: Standard mass loadings applied for the POC evaluation.

Description	Pu-239/238 Mass (grams)
High Mass MT-52	135.9
Low Mass MT-52	14.2
High Mass MT-83	5.61
Low Mass MT-83	0.25

Table 4 describes the results from the neutron evaluation and Table 5 describes the results for the gamma modality.

Table 4: Neutron results for standard loading of the empty POC container.

Standard Loading	Average Value Pu-240 eff mass (g)	%R	STDEV	%RSD
NTP-0140: 15 grams: MT-52	0.892	97.7%	0.007	0.83
NTP-0146, 0156,0164: 145 grams: MT-52	9.283	105.5%	0.155	1.67
NTP-0239: 0.3 grams: MT-83	0.690	105.1%	0.027	3.87
MF40F: 7.6 grams: MT-83	16.026	111.4%	0.379	2.36

The neutron results showed a small bias for the standard POC drums. The largest average percent recovery value was for the 7 gram MT-83 standard loaded in the empty POC drum. The percent relative standard deviation %RSD values would meet WIPP requirements. The upper limit for the %RSD value quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 (DOE/WIPP-02-03122[2]) for six measurements is 14%. The recovery values for the standard measurements would also meet the WAC criteria of 30 % for calibration verification measurements. The neutron results were based on standard neutron coincidence counting techniques.

Table 5: Gamma results for standard loading of the empty POC container.

Standard Loading	Average Value Pu239/Pu238 Mass (g)	%R	STDEV	%RSD
NTP-0140: 15 grams: MT-52	21.040	148.6%	0.273	0.83
NTP-0146, 0156,0164: 145 grams: MT-52	169	124.2%	21.57	12.78
NTP-0239: 0.3 grams: MT-83	0.291	116.3%	0.014	4.97
MF40F: 7.6 grams: MT-83	5.715	101.9%	0.102	1.78

The gamma results showed a larger bias for the standard POC drums than the neutron results. The largest average percent recovery value was for the 15 gram MT-52 standard loaded in the empty POC drum. The percent relative standard deviation %RSD values would meet WIPP requirements for each measurement. The upper limit for the %RSD value quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 (DOE/WIPP-02-03122[2]) for six measurements is 14%. The recovery values for three of the four standard measurements would also meet the WAC criteria of 30% for calibration verification measurements. The 15 gram standard measurement would not meet the accuracy requirements in the WAC. The gamma results were evaluated based on direct gamma quantification of Pu-238 or Pu-239 depending on the material type applying a self-absorption correction. Self- absorption correction was not applied to the standard MT-83 POC containers. Weighted average values were used to calculate the mass values for Pu-238 and Pu-239 using the 766 keV and 786 keV gamma-ray energy lines.

POC CALORIMETRY COMPARISON RESULTS

After the waste and standard comparison a third comparison was performed to determine the viability of measuring POC's with the current calibration. For this comparison, five items were measured by calorimetry plus gamma-ray isotopic measurements, then placed in POCs and moved to HENC #2 for evaluation. Five different loadings were evaluated for the calorimetry characterized POCs. Table 6 contains a summary of the data for the neutron evaluation and Table 7 contains the data for the gamma modality. A total of six measurements were made for each POC.

Table 6: Neutron results for waste loading of POCs evaluated by calorimetry.

Calorimetry Results	Average Value Pu239/Pu238 Mass (g)	%R	STDEV	%RSD
92.2 grams MT-52	111.1	121	6.3	5.6
117.5 grams MT-52	129	110	9.5	7.4
5.5 grams MT-83	5.1	93	0.3	5.1
12.4 grams MT-83	11.3	91.2	0.2	1.9
19.6 grams MT-83	16.7	85	0.4	2.4

The neutron results showed a small bias for the calorimetry characterized POC drums. The largest average percent recovery value was for the 117.5 gram MT-52 calorimetry characterized POCs. The percent relative standard deviation %RSD values would meet WIPP requirements. The upper limit for the %RSD value quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 (DOE/WIPP-02-03122[2]) for six measurements is 14%. The recovery values for the standard measurements would also meet the WAC criteria of 30 % for calibration verification measurements. The neutron results were based on standard neutron coincidence counting techniques.

Table 7: Gamma results for waste loadings of POCs evaluated by calorimetry.

Calorimetry Results	Average Value Pu239/Pu238 Mass (g)	%R	StDev	%RSD
92.2 grams MT-52	112	121	2.3	2.0
117.5 grams MT-52	126	107	3.4	2.7
5.5 grams MT-83	4.89	89	0.2	3.3
12.4 grams MT-83	9.58	77.3	0.1	1.5
19.6 grams MT-83	15.0	77	0.3	1.7

The gamma results showed a larger bias for the standard POC drums than the neutron results specifically for the MT-83 containers. The largest average percent recovery value was for the 19.6 gram MT-83 standard loaded in the empty POC drum. The percent relative standard deviation %RSD values would meet WIPP requirements for each measurement. The upper limit for the %RSD value quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 (DOE/WIPP-02-03122[2]) for six measurements is 14%. The recovery values for the three of the four standard measurements would also meet the WAC criteria of 30 % for calibration verification measurements. The gamma results were evaluated based on direct gamma quantification of Pu-238 or Pu-239 depending on the material type applying a self-absorption correction. Self- absorption correction was not applied to the standard MT-83 POC containers. Weighted average values were used to calculate the mass values for Pu-238 using the 766 keV and 786 keV gamma-ray energy lines.

ISOTOPIC MEASUREMENTS

Isotopic measurements were also performed during the evaluation of the POC containers. Two different isotopic programs were evaluated, MGA and FRAM. MGA version 9.63F is embedded in the counting system and the analysis is performed at the time of the measurement. The FRAM analysis is performed upon reanalysis of the gamma result.

The MGA isotopic program embedded in the HENC#2 software was not able to determine the isotopic composition of the drums because of the highly attenuated gamma signal. However, a later isotopic analysis of drums with higher plutonium loadings using the FRAM program was successful in analyzing the composition. Unfortunately, the statistical uncertainty in the Pu-240 isotopic result was consistently large, even after measurements of two hours duration. This large statistical uncertainty, in turn, increased the Pu-239 Fissile Gram Equivalent (FGE) total measurement uncertainty and thereby increased the rejection potential for drums containing greater than 200 FGE. FRAM was not able to analyze waste drums containing less than approximately 25 g of MT-52 and did not credibly analyze the Heat Source waste drum.

CONCLUSIONS

The test results indicate that container effects from the 12" pipe and fiberboard dunnage do not impose undue difficulty for the HENC neutron measurements of POC standard drums. The gamma-ray measurements for the POC standard drums indicated that the HENC#2 can measure higher mass ^{238}Pu drums. The tests also determined that the POC drums are within the gamma bulk density matrix calibration and neutron Add-a-Source correction ranges for the HENC. This is borne out by the accuracy and precision of the HENC measurements of standards in an otherwise empty POC drum. For both the standards and the waste drum measurements, the self absorption correction method results were biased high. In addition, the isotopic measurements either could not be performed or had large errors for both the waste and standards measurements.

The neutron and self absorption corrected gamma ray measurement of the WG POC waste drums do not provide satisfactory agreement with baseline values established by the waste generators. The cause of the disagreement is not fully understood. However, after adding the calorimetry data to the comparison, the HENC values compared quite well. Since the POC test measurements passed two out of the three comparisons, it was determined that the current calibration could be used for these types of containers. It was determined that the original waste values used for the first comparison were not a reliable as the calorimetry data used for this evaluation.

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