Evaluation of Nondestructive Assay Characterization Methods for Pipe-Over-Pack Containers

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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 or POC's exhibit 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. Also, chemical and matrix properties of the Pu contaminated waste in the POCs that may affect the measurement result are generally unknown in advance of the measurement. 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.

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 instrument, a High Efficiency Neutron Counter (HENC) with an integrated high purity germanium detector incorporates both passive neutron and high resolution gamma counting capabilities. 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 matrix and source material and assays the drum with a calibration based on the known density of the matrix. Both methods are supplemented by a simultaneous gamma isotopic measurement using Multi-Group Analysis (MGA) software to determine the plutonium isotopic composition. If MGA fails to provide a viable isotopic result Fixed energy Response function Analysis with Multiple efficiencies (FRAM) could be used to replace MGA results. Acceptable knowledge (AK) may also be used in certain instances.

This report will discuss the two measurement methods in detail for POC analysis. 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 safeguards data. Empty POCs loaded with known sources have also been measured to determine the viability of the technique. Finally, a brief discussion of the conclusions that can be drawn from the tests will be offered.

INTRODUCTION

Nondestructive assay techniques measure radiation emitted from nuclear materials without altering their physical or chemical state. At Los Alamos National Laboratory there are a number of Pipe OverPack containers (POC's) 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 POC's 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 methods used to measure POC's at Los Alamos National Laboratory (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 and matrix effects on the measurements.

PIPE OVERPACK

The standard pipe overpack identified in CH-TRAMPAC Revision 2 was used for the test measurements. There are two types of POC's 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. The following figures show the actual configuration of the POC.

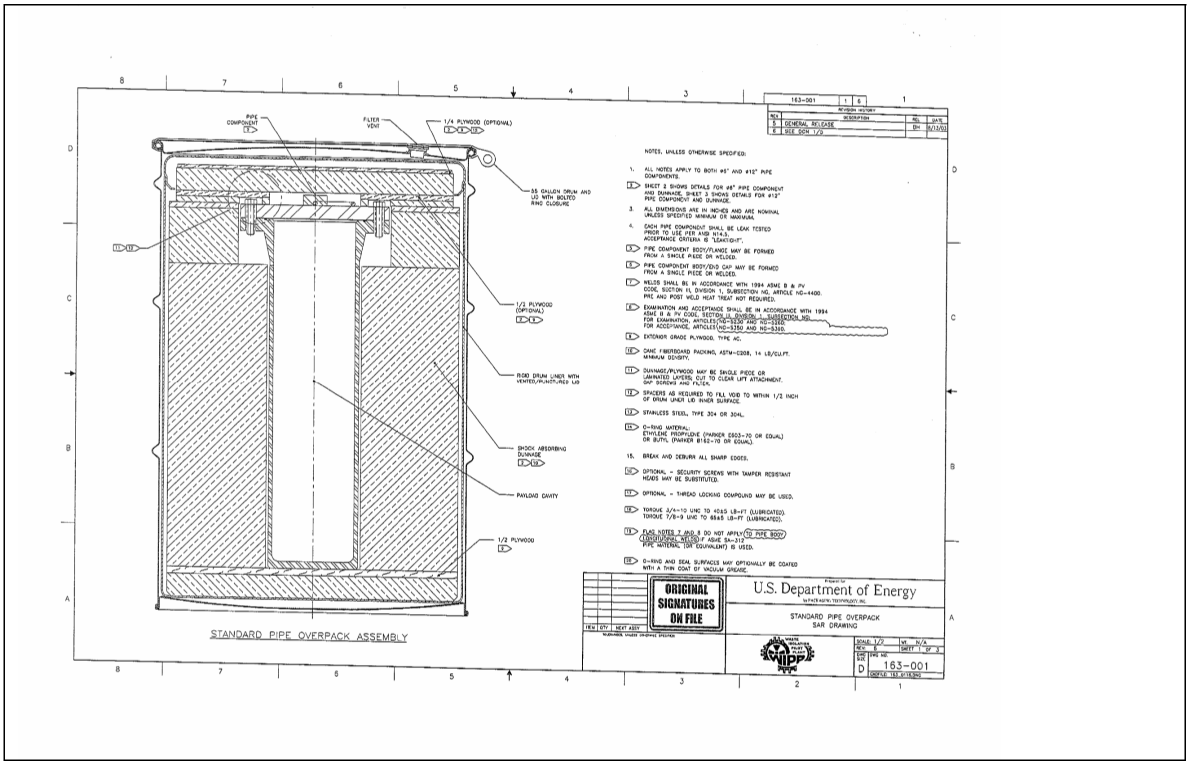

Figure 1: Standard pipe overpack assembly as shown in the CH-Trampac Rev. 2 [1]. All notes in the diagram apply to the 6 inch and 12 inch pipe overpack containers.

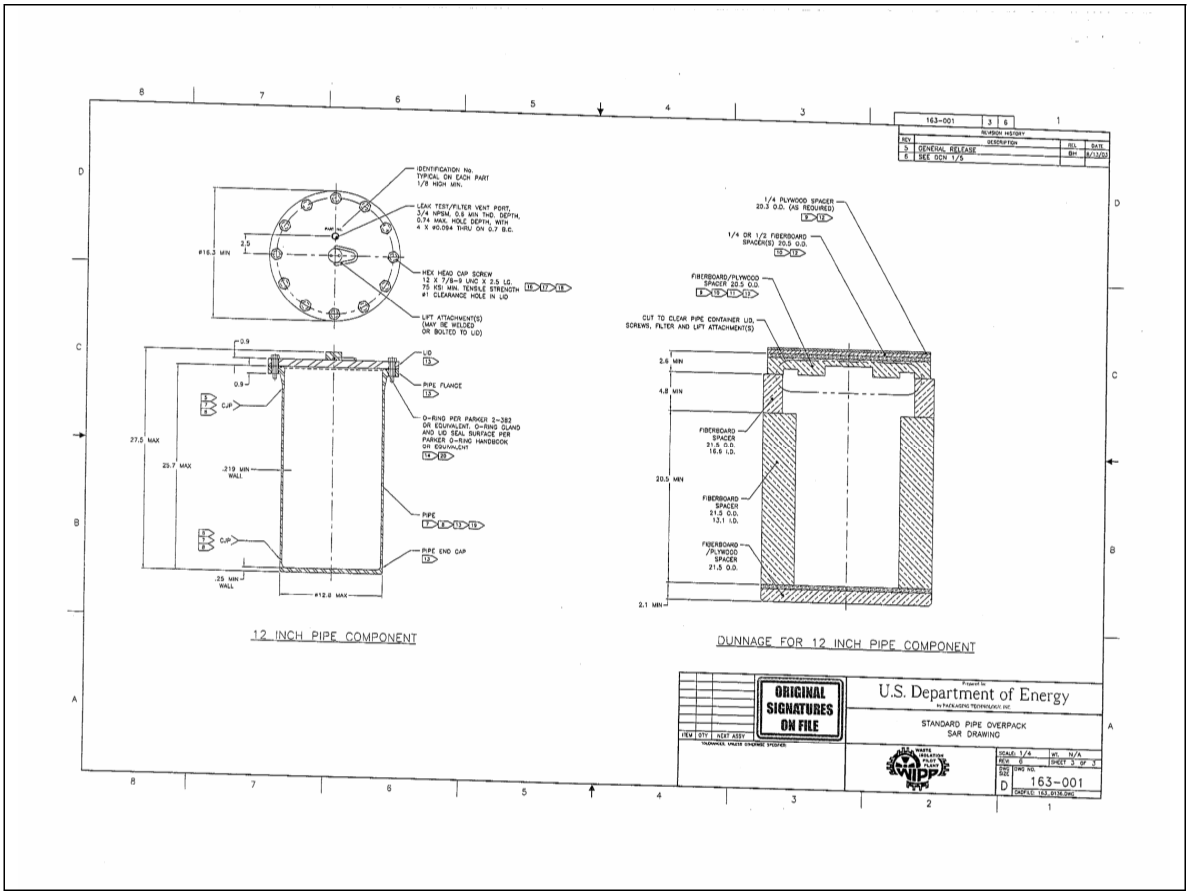

Figure 2: 12 inch pipe component from the CH-TRAMPAC Rev. 2 [1].

The waste placed in POC containers was produced at TA-55. 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 and 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 MCS 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 1400 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,, and sand. The densities of the matrix drums range from 0.002 g/cc to 1.64 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.

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 (MGA) software under the control of NDA 2000. This measurement is performed concurrently with the quantitative gamma ray measurement. Fixed energy Response function Analysis with Multiple efficiencies (FRAM) software may also be applied during data review.

POC EVALUATION DESCRIPTION

The POC evaluation consisted mainly of two different comparisons. The first was comparing TA-55 safeguards measurements of actual POC waste with HENC results. The second portion of the evaluation was to load an empty POC container with known standards and to determine the performance based on WIPP criteria.

The POC waste evaluation consisted of 4 different containers. The first container was a low 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 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 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 values were either determined by neutron or gamma-ray NDA instruments. However the 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 consisted of 1 container. 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 1 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.

Table 1: 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

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 TA-55 baseline values and the standard values are expressed as % Recovery (%R).

POC WASTE EVALUATION RESULTS

The first tests performed on the HENC were the waste POC containers from TA-55. The data obtained from the HENC assays was compared to safeguards data from TA-55. The following table describes the neutron results for the waste POC container.

Table 2: Neutron results from POC TA-55 waste measurements. The values stated in this table are the average of six standard 1800 second measurements. The asterisk for drum #61741 designates that the mean value is from only five measurements.

Waste Drum	Average Pu Mass Neutron	% Recovery	STDEV	%RSD
2.6 Gram MT-52 (61741*)	3.9	151.3%	0.5	13.4
47.2 Gram MT-52 (63446)	95.0	201.2%	12.1	12.7
111 Gram MT-52 (63424	145.8	131.4%	6.9	4.7
5.1 Gram MT-83 (62299)	8.6	168.3%	1.7	2.0

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 3: Gamma results from POC TA-55 waste measurements. The values stated in this table are the average of six standard 1800 second measurements. The asterisk for drum #61741 designates that the mean value is from only five measurements.

Waste Drum	Average Pu Mass Gamma	% Recovery	STDEV	%RSD
2.6 Gram MT-52 (61741*)	4.3	164.9%	0.4	9.0
47.2 Gram MT-52 (63446)	82.7	175.1%	4.1	5.0
111 Gram MT-52 (63424	145.2	130.8%	3.7	2.5
5.1 Gram MT-83 (62299)	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 following table describes the results from the neutron evaluation.

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 the 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 an Pu-239 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 Multi-Group Analysis 9.63F (MGA), and Fixed energy Response function Analysis with Multiple efficiencies (FRAM). MGA version 9.63F is embedded in the counting system and the analysis is performed at the time of the measurement.

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. For this reason, we conclude that AK isotopic values, when established and credible, are preferable to measured values if the Pu-240 uncertainty is large (> 20%). FRAM was not able to analyze waste drums containing less than approximately 25 g of WG Pu and did not credibly analyze the Heat Source waste drum (62299).

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 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 were either not 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 generators. Both the neutron and gamma ray results indicate a high bias. That bias was not substantially improved by the use of truncated or full multiplicity neutron methods or by the weighted average of high energy Pu-239 gamma rays. The cause of the disagreement is not fully understood. It may be due to incorrect baseline values. It is also possible that it is caused by plutonium or matrix effects in the waste or by the presence of plutonium salts with concentrations of PuF_4. If a material fraction of the waste has lumps or is complexed as a salt, there are no existing NDA techniques that have a proven capability to measure the drummed POC waste. The material could be measured by calorimetry while it is in smaller containers. However, drum calorimeters are not available at the waste measurement facility at Los Alamos National Laboratory. Also, calorimetric measurements of drummed waste are still in an experimental stage. MT 83 POC waste, on the other hand, was measured with acceptable accuracy and precision by the quantitative gamma method when the weighted average of the 766 keV and 786 keV lines from Pu-238 were used.

If the baseline values are substantially correct, it may be possible to develop specialized calibration correction factors for weapons grade POC waste drums. The factors could be based on an averaged difference between the HENC and baseline values. However, before such factors can be developed, the number and expected frequency of the different waste forms (for example, the type of salt, presence of PuF_4, possibility of large amounts of Am-241, etc) that generated the waste and the credibility of the baseline values must be established. More than one calibration correction factor may be needed.

REFERENCES

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