Evaluation of 55-gallon Lead-Lined Calibration for the Gamma Modality of the High Efficiency Neutron Counter

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ABSTRACT

Nondestructive assay (NDA) measurements of Transuranic (TRU) waste at Los Alamos National Laboratory (LANL) packed in Lead Lined 55-gallon drums are associated with a number of complexities. Some of the debris waste or lead-lined drums containing cans will be repackaged into non-lead lined containers. However a number of the lead lined drums contain a concrete matrix. The ability to remove either the concrete matrix or the lead liner from the drum is very limited. Currently there are a number of lead lined containers at LANL that require evaluation for shipment to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, NM.

At LANL, one of the High Efficiency Neutron Counters has been used to determine the appropriateness of both passive neutron and quantitative gamma ray methods for measuring lead lined drums. 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 isotopic measurement using Multi-Group Analysis (MGA) to determine the plutonium isotopic composition. If MGA fails to provide a viable isotopic result, then an alternative isotopic determination is made using Fixed-energy Response Function Analysis with Multiple efficiencies (FRAM) software. Isotopic distributions derived from process knowledge may also be used in certain instances.

This report discusses the two methods in detail as they pertain to lead-lined drum measurements. Included in the discussion is a description of the setup parameters and calibration techniques for the instrument. The gamma modality was calibrated using lead lined calibration drums. The neutron modality, calibrated with Pu sources in non-lead lined containers, will be evaluated using the current Add-A-Source matrix correction technique. The discussion will include confirmation of the calibration for WIPP Waste Acceptance Criteria and calibration validation data. The conclusion discusses whether the new gamma calibration meets the WIPP Waste Acceptance criteria and whether the current neutron calibration is viable.

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 lead-lined containers containing large amounts of nuclear material that are assayed with NDA systems. It is an important part of the Transuranic Waste Program at LANL to remove these high activity waste drums from the site.

Measuring transuranic waste placed in lead lined containers poses a number of technical issues for NDA. The lead liner causes a large amount of attenuation for the gamma-rays emitted from the drum. The lead lined drum population consists of lead lined drums with cemented waste. Some of the drums contain debris waste but the plan is to repackage this waste at some point in time. Cemented waste contains hydrogen that can reduce the effectiveness of the neutron modality of the system.

The purpose of this report is to describe the effectiveness of the new gamma calibration used to measure lead-lined drums at Los Alamos National Laboratory (LANL) in detail. Included in the discussion will be descriptions of the lead-lined containers, instrument setup parameters, calibration techniques and analysis restrictions for the methods. A description of confirmation measurements used to validate the gamma calibration will be discussed.

LEAD LINED CONTAINERS

The lead lined containers were used by Los Alamos to reduce the radiation dose to workers from high activity waste. A standard 55-gallon drum was lined with lead with a nominal thickness of 1/16 of an inch. The sides of the drum were lined and a lead disc was placed in the bottom of the drum. The concrete and waste was then added to the drum and a lead disc was placed on top of the waste. The drum lid was then placed on the drum and sealed. Historically these drums are known to contain high levels of ^{241}Am so a new gamma calibration was required to proper quantify the Am-241 content under these conditions. Also the lead liners cause a large amount of attenuation on the gamma modality of the system.

NDA METHODS USED TO MEASURE LEAD-LINED CONTAINERS

The LANL HENC (HENC #2), shown in Figure 1, 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 1: 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 plutonium 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. Lead lined calibration drums were manufactured to calibrate only the gamma modality of the HENC's.

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 1228 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 lead lined calibration drums range from 0.03 g/cc to 1.79 g/cc. The density calculation do not contain the weight of the lead. 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.

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 ^{241}Am 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.

CALIBRATION MATERIALS

Gamma-emitting reference materials

The gamma ray efficiency calibration that relates count rates to decay rates was performed with a set of six NIST-traceable gamma ray reference sources. These working reference materials are North American Scientific Inc. Model ENV4057 mixed gamma line reference sources, with a nominal combined activity of 60 μCi (30 μCi each of ^{241}Am and ^{152}Eu). The radioactive materials are dispersed in epoxy, and are encapsulated in aluminum tubes. In conjunction with the Sn/Cd filter, the emitted gamma rays provide an effective calibration range from 122 keV to 1228 keV, suitable for measuring the radionuclides anticipated in LANL waste. These reference sources are traceable to the national standards base.

Reference drums were assembled using a set of six gamma ray reference line sources in five different matrix drums that were lead-lined. The five lead-lined matrix drums used for the calibration were an empty (0.03 g/cm^3), Homasote® (0.51 g/cm^3), particle board (0.66 g/cm^3), sand (1.49 g/cm^3), and concrete (1.79 g/cc). These matrices provided an operational density range from 0.03 g/cm^3 to 1.79 g/cm^3. The reference drums have nine plastic tubes, in a spiral configuration such that the tubes are all different distances from the drum axis. Figure 1 shows the tube configuration for the matrix drums. For each matrix drum, the line sources were placed in order of increasing serial number in tubes 2, 3, 4, 5, 6, and 9 respectively. The gamma ray reference sources and their tube positions are described in Table 1, and shown in Figure 1.

Table 1: Gamma calibration reference sources.

Source ID (S/N)	Nuclide	Half Life (y)	Activity (μCi)	Activity Ref. Date
63723 (Tube Position 2)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.756 ± 1.35%[1] 5.682 ± 1.36%	04/01/2005
63724 (Tube Position 3)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.284 ± 1.35% 5.794 ± 1.36%	04/01/2005
63726 (Tube Position 4)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.523 ± 1.35% 5.581 ± 1.36%	4/01/2005
63727 (Tube Position 5)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.606± 1.35% 5.408 ± 1.36%	04/01/2005
63728 (Tube Position 6)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.854 ± 1.34% 6.094 ± 1.34%	04/01/2005
63960 (Tube Position 9)	^{241}Am ^{152}Eu	432.7 ± 0.5 13.33 ± 0.04	5.915 ± 1.36% 5.599 ± 1.39%	04/01/2005

*** Half-life for ^{152}Eu from Brookhaven National Nuclear Data Center (USDOE).**
[1] The uncertainty values shown are at 1-sigma.

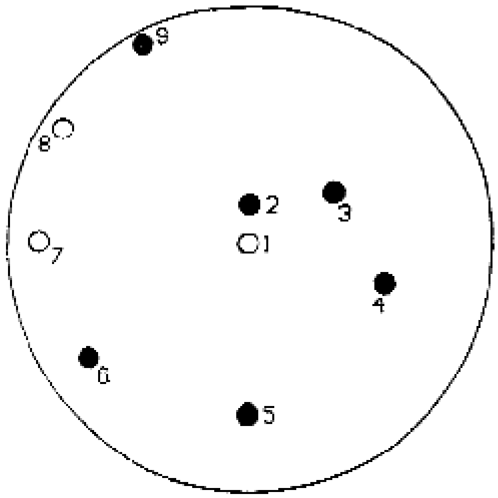

Figure 1: Top View, Cross Section of Calibration Drums. Figure 1 shows the positions of the line sources for the empty, homasote, particle board, sand drum and concrete drum.

LEAD-LINED WASTE MEASUREMENTS

Measurements were performed on nine 55-gallon containers that were packaged with waste. The drums were lead-lined and there was Acceptable Knowledge (AK) information available for each drum. The AK contained the amount of total Plutonium, Americium and the Plutonium isotopic mix for the drum. The measurement results for the nine drums are shown in Table 2 applying quantitative gamma data. The neutron results for these drums are shown in Table 3.

Table 2: Lead-lined calibration results using quantitative gamma data. MT refers to the type of Plutonium or Material Type. WG refers to Weapons Grade Plutonium.

Drum ID	MT	Density (g/cc)	Baseline Total Pu (g)	Measured Total Pu (g)	Pu %R	Baseline Total ^{241}Am (g)	Measured ^{241}Am Mass (g)	^{241}Am %R
1	WG	2.05	86	83.9	97.6%	7.2	6.48	89.95%
2	WG	1.98	34.1	34.5	101.1%	8.7	6.85	79.0%
3	WG	1.98	17.6	20.7	117.4%	6.4	6.57	102.5%
4	WG	1.34	61.4	58.5	95.3%	N/A	6.28	N/A
5	WG	1.15	8.23	12.4	150.7%	N/A	9.94	N/A
6	WG	1.24	92.32	61.7	66.8%	N/A	6.34	N/A
7	WG	1.18	29.00	29.4	101.4%	N/A	3.34	N/A
8	WG	1.94	71.3	77.9	109.3%	7.5	6.99	93.2%
9	WG	2.03	40.0	44.9	112.3%	8.3	7.50	90.36%

Table 3. Summary of HENC 2 neutron results for the lead-lined drums.

Drum ID	MT	Density (g/cc)	Baseline Total Pu (g)	Measured Total Pu (g)	%R
1	WG	2.05	86.0	138.0	160.46%
2	WG	1.98	34.13	65.00	190.45%
3	WG	1.98	17.60	26.60	151.36%
4	WG	1.34	61.40	159.0	258.96%
5	WG	1.15	8.23	55.60	1076.55%
6	WG	1.24	92.32	149	161.39%
7	WG	1.18	29.00	52.80	182.07%
8	WG	1.94	71.3	125	175.32%
9	WG	2.03	40.00	81.00	202.50%

Overall the gamma measurements for the waste drums were acceptable. The neutron measurements all overestimated due to high add-a-source corrections. Drum #5 showed a high gamma recovery and drum #6 showed a low recovery and currently there is no explanation. A possible explanation could be that the AK information could be incorrect for these drums but it is more likely due to the fact these are not concrete drums, but contain individual cans of material that are not uniformly distributed in the drum. These drums would not be measured on the systems until a viable Total Measurement Uncertainty (TMU) was developed. The decision was made to move ahead with the gamma data and only use neutron results if the add-a-source correction is in an acceptable range.

LEAD-LINED 55-GALLON WIPP REQUIRED CONFIRMATION RESULTS DESCRIPTION

In order to determine that a new calibration of an NDA instrument is correctly established, DOE/WIPP-02-3122 Appendix A requires that the accuracy and precision of the instrument be confirmed by performing replicate measurements of Pu reference sources in non-interfering matrices. The reference sources used for the calibration confirmation are certified reference materials, or working reference materials traceable to the national standards database. The confirmation reference sources must be different from the ones used for calibration, and approximately span the range of expected operation. Only the gamma modality for this system was re-calibrated for the lead lined matrix. The data reported for the neutron confirmation is from a previous calibration.

The lead lined gamma calibration was confirmed by measuring Plutonium sources in a standard zero-matrix drum. Each drum was measured six times with a total of forty eight measurements in all. The drum configurations are described in Table 4.

Table 4: Configuration of standard drums for calibration confirmation. The Pu sources are independent of the sources used for calibration as required by the WAC.

Standard ID	Pu Mass (g)	Standard Matrix	Position of Standards in Drum	Simulated or Actual Waste Matrix	Number of Measure-ments
NTP-085 NTP-0099	**0.60**	Diatomaceous Earth	Tube 2 Ht: 15" Tube 3 Ht: 9"	Zero Matrix (PDP Drum 001)	6
PDP1-3.0	**3.02**	Diatomaceous Earth	Tube 2 Ht: 15" Tube 3, Ht: 9"	Zero Matrix (PDP Drum 001)	6
NTP-0140	**15.048**	Diatomaceous Earth	Tube 3, Ht: 15"	Zero Matrix (PDP Drum 001)	6
NTP-0140 NTP-0148 NTP-0156 NTP-0164	**160.00**	Diatomaceous Earth	Tube 1, Ht: 15" Tube 3, Ht: 9" Tube 2, Ht: 3" Tube 2, Ht: 21"	Zero Matrix (PDP Drum 001)	6
NTP-0164 NTP-0156 NTP-0148 NTP-1040 NTP-0007 PDP 40.0 Pu1 PDP1-10	**217.6**	Diatomaceous Earth	Tube 1, Ht: 1" Tube 1, Ht. 21" Tube 2, Ht. 10" Tube 2, Ht. 20" Tube 3, Ht. 10" Tube 3, Ht. 20" Tube 1, Ht. 11"	Zero Matrix (PDP Drum 001)	6

Standard ID	Pu Mass (g)	Standard Matrix	Position of Standards in Drum	Simulated or Actual Waste Matrix	Number of Measure-ments
&NTP-0239 (Pu-238)	**0.306**	Diatomaceous Earth	Tube 2, Ht. 15"	Zero Matrix (PDP Drum 001)	6
&MF40F (Pu-238)	**7.234**	Diatomaceous Earth	Tube 2, Ht. 15"	Zero Matrix (PDP Drum 001)	6
*MF-22 (Pu-238)	**10.82**	Diatomaceous Earth	Tube 2, Ht: 10"	Zero Matrix (PDP Drum 001)	6
*MF-22 *MF-40F (Pu-238)	**18.0**	Diatomaceous Earth	Tube 2, Ht: 4" Tube 2, Ht: 15"	Zero Matrix (PDP Drum 001)	6

Mass loadings of 0.6 g, 3 g, 15 g, 160 g and 217.6 g Pu were selected as representative of typical waste drums for multiplying waste forms (Weapons grade Pu or predominately ^{239}Pu). Mass loadings of 0.3 g, 7 g, 10.82 g and 18.00 g Pu were selected as representative of typical waste drums for non-multiplying waste forms (Heat source Pu or predominately ^{238}Pu). The measurements were all performed as Item Counts following the procedure to be used for "unknown" waste drums. The confirmation measurement results are listed in Table 3. Following established practice, if viable measured isotopic fractions are available, they are used. Otherwise, the declared or certificate isotopic fractions are used. The statements pertaining to multiplying and non-multiplying waste forms describe neutron interactions.

The confirmation measurement results were evaluated against the criteria specified in DOE/WIPP-02-3122 Appendix A, Table A-3.2 (repeated in CCP-TP-064). Accordingly, the accuracy of a measurement (percent recovery or %R) must be >70% and <130%, and the precision (%RSD) must be within 14% for six replicate measurements. Based on these criteria, accuracy (%R) and precision (%RSD) were acceptable for all weapons grade drums, for the gamma modalities. All of the confirmation runs using Pu-238 sources also passed the WAC requirements. For this report only the Gamma results are shown in Table 5.

Table 5: Summary of confirmation measurement results using the lead-lined efficiency curve for the gamma modality only.

Confirmation Standard IDs	Average $^{239/8}$Pu Mass (g)	%R and %RSD
NTP-0085 NTP-0099	0.78	%R = 138.05 %RSD = 2.10
PDP1-3.0	2.83	%R = 116.3% %RSD = 2.54%
NTP-0140	14.1	%R = 113.2% %RSD = 1.10%
NTP-0140 NTP-0148 NTP-0156 NTP-0164	150.04	%R = 89.2% %RSD = 1.29%
NTP-0164 NTP-0156 NTP-0148 NTP-1040 NTP-0007 PDP 40.0 Pu1 PDP1-10	204	%R = 82.48% %RSD = 0.65%
NTP-0239 (^{238}Pu)	0.25	%R = 97.6% %RSD = 4.78%
MF40F (^{238}Pu)	5.64	%R = 102% %RSD = 2.94%
MF-22 (^{238}Pu)	8.2	%R=100.3 %RSD=2.47
MF-22 MF-40F (^{238}Pu)	13.8	%R=87.4 %RSD=1.35

Another set of six measurements was also performed to validate the lead-lined calibration in an interfering matrix. A 50 gram weapons grade standard was placed in a lead-lined concrete drum to validate the response of the efficiency curve at this density. The standard was placed in Tube 3 at a height of 15 inches (T3H15) for the validation measurement. The results for these measurements are shown in Table 6.

Table 6: Summary of validation measurement with lead lined concrete matrix drum.

Validation Set	Average ^{239}Pu Mass (g)	%R and %RSD
1 RANT-50-1	46.5	%R=120.0
		%RSD=1.03

The validation measurement results were evaluated against the criteria specified in DOE/WIPP-02-3122 Appendix A, Table A-3.2 (repeated in CCP-TP-064). Accordingly, the accuracy of a measurement (percent recovery or %R) must be >70% and <130%, and the precision (%RSD) must be within 14% for six replicate measurements. The validation measurements met these requirements.

ISOTOPIC MEASUREMENTS

Plutonium isotopic measurements were not available for most of the drums used to confirm the calibration. Some of the high energy lines can be used for isotopic calculations but currently the plan is to use Acceptable Knowledge for the isotopic information.

CONCLUSIONS

The test results indicate that the lead lined calibration is viable under WIPP Waste Acceptance Criteria. The confirmation results met the WIPP requirements for precision and accuracy. Due to the lead liner and the strength of the source the spectral data will only be viable from 244 keV and above. The neutron results will be used only when the matrix correction results are within acceptable parameters.

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