Upgrade and Certification of the Los Alamos National Laboratory SHENC 2011 - 12270

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ABSTRACT

Nondestructive assay measurements of Transuranic (TRU) waste at Los Alamos National Laboratory (LANL) required the addition of a standard waste box (SWB) assay system. A Super High Efficiency Neutron Counter (SHENC) located at Hanford was identified to be relocated to LANL. After careful evaluation of waste streams at LANL, it was determined that the current configuration of the SHENC was not sufficient to quantify certain waste streams.

At LANL, there is still a large amount of waste that needs to be retrieved and re-packaged within SWB's to meet agreements with the State of New Mexico. Prior to relocating the SHENC, the only assay systems available were High Efficiency Neutron Counters having only a 55-gallon drum capacity. Further analyses indicated that the SHENC system should be capable of quantitative gamma measurements that are to be linked, and combined, with the neutron measurements. The SHENC system was therefore augmented with a new high-resolution gamma spectroscopy system using BE5030 detectors and upgraded gamma electronics. The neutron side of the system was also upgraded with an advanced shift register (JSR-15), an improved Programmable Logic Controller and NDA-2000 software.

This report will include calibration of both the neutron and gamma modalities of the SHENC system and how the modality results are combined to produce a single assay result. Preliminary performance results will be discussed based on both mock and real waste measurements. Discussions will also include a complete description of the adjustable parameters as well as the calibration plan, techniques and validations including calibration confirmation based on the Waste Isolation Pilot Plant Waste Acceptance Criteria (WIPP-WAC).

INTRODUCTION

Nondestructive assay techniques measure radiation emitted from nuclear materials without altering their physical or chemical state. Nondestructive assay techniques can be used on a variety of containers or objects. At Los Alamos National Laboratory (LANL) there are a number of objects that require packaging in a Standard Waste Box (SWB). Previously at Los Alamos, the only Waste Isolation Pilot Plant (WIPP) certified NDA instruments were designed for 55-gallon drums. Other DOE sites such as Rocky Flats, Hanford, and Idaho have used WIPP certified box counters extensively.

The purpose of the SHENC upgrade at Los Alamos was to provide a standardized and updated method for NDA. The first issue was to add a quantitative gamma component to the instrument. The variety of waste streams at Los Alamos can cause significant technical issues for neutron based measurement techniques. To help minimize the amount of rejected SWBs two High Purity Germanium (HPGEs) Detectors were added to the system. With the addition of the HPGEs, the current software used on the SHENC could not efficiently combine both gamma and neutron data. The system was updated with NDA 2000 software to not only provide an effective approach to combining the data but also to standardize the software used for WIPP certified measurements at Los Alamos.

With the addition of new software a new Programmable Logic Controller (PLC) was added to the system to provide an interface between the Add-A-Source and NDA-2000. With the new PLC, the Add-A-Source could then be controlled by NDA-2000.

Other components on the system were upgraded as well. Since new HPGE's were added to the system new integrated multi-channel analyzers were also added to the SHENC. The neutron side of the system was upgraded with a JSR-15 shift register that could be replaced easily if the unit failed.

The purpose of this report will be to discuss the system upgrade and the calibration of the system for WIPP certification at Los Alamos.

NDA METHODS USED TO MEASURE SOLID WASTE BOXES

The Los Alamos SHENC shown in Figure 1, is a hybrid NDA instrument with two HPGEs for gamma analysis and a separate chamber for neutron measurements. The original SHENC design did not have the capability for full quantitative gamma assay. With the added capability, the system integrates the two NDA counting modes: passive neutron and gamma-ray assay.

Figure 1: Super High Efficiency Neutron Counter at Los Alamos National Laboratory.

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

The SHENC passive neutron counter employs neutron coincidence and multiplicity counting of Standard Waste Boxes. The counter utilizes 260 gamma-hardened He-3 proportional detectors, divided into 52 detector banks, arranged in a 4-π geometry about the assay cavity. Each tube contains He-3 with a fill pressure of ten atmospheres. The nominal assay cavity is 55 inches wide by 71 inches long by 36 7/8 inches 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

$$m_{Pu-240Eff} = 2.52m_{Pu-238} + 1.0m_{Pu-240} + 1.68m_{Pu-242} \qquad \text{(Eq. 1)}$$

The calibration curve is used to calculate a Pu-240 effective mass value for neutron measurements of waste items. 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 Solid Waste Boxes.

GAMMA-RAY ASSAY MODALITY

The gamma portion of the SHENC system uses two Broad Energy Germanium (BEGe) gamma-ray detectors. The detectors are mounted on the loadside of the counter perpendicular to, and pointing towards, the vertical axis of the sample box on the gamma side rotator. Each detector is mounted such that it can be withdrawn from the counter when required. The detector is firmly positioned in the system upon reassembly for routine gamma ray assays. The configuration of the detectors can be seen in Figure 2. Collimation was added to the Gamma modality as well. The collimation allows for each detector's field-of-view to contain the entire SWB.

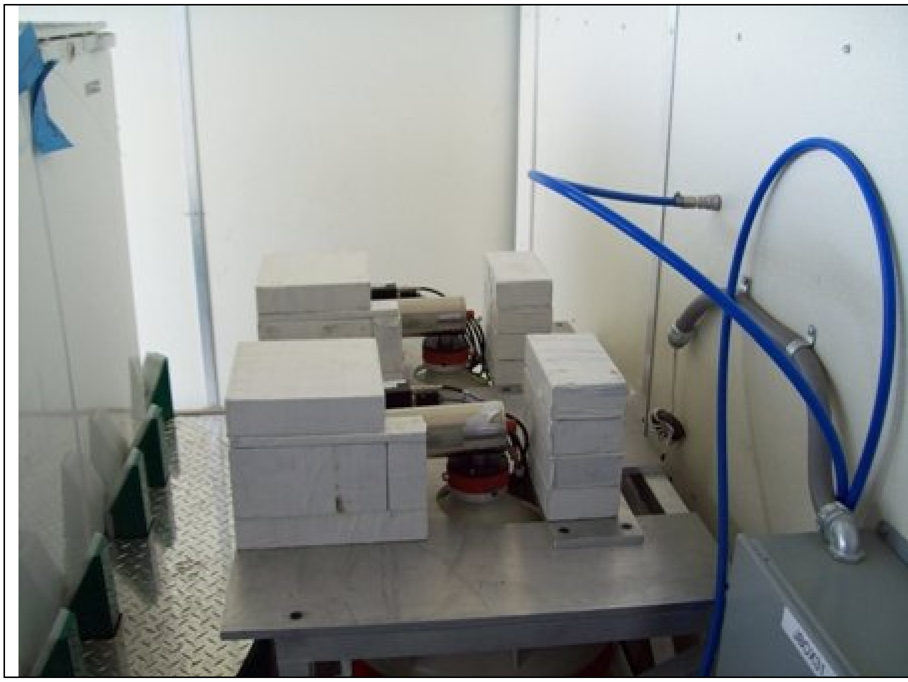

Figure 2: HPGE configuration for the Los Alamos SHENC.

The spectra from the BEGe gamma detectors are 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 1600 keV as a function of the density of the matrix material. Five matrix boxes were used to establish the efficiency calibration: Zero Matrix, Cardboard, Cardboard/Plywood, Cardboard/High Density Poly/Plywood, and Plywood/Low Density Poly. The densities of the calibration containers range from 0.00 g/cc to 0.788 g/cc. Am-241/Eu-152 line sources that extend the full axial length of the boxes are used for the calibration. The sources are placed in a configuration such that, when the box was 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 detectors are used to measure the plutonium isotopic composition of the waste during each box 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 may also be applied when required.

NEUTRON CALIBRATION DESCRIPTION

The passive neutron mass calibration that relates coincidence count rates to Pu-240$_{eff}$ content was performed in 2011. Many of the parameters used in the setup of the SHENC were taken from References [1], [2], and [3].

The calibration of the neutron counter used Plutonium sources from Los Alamos National Laboratory. These reference materials were fabricated at Los Alamos National Laboratory and are traceable to the national standards database. Their characteristics are described in Table 1, and the source certificates are described in Reference [4]. The sources were positioned in the tubes denoted on Figure 3.

Table 1: Pu reference sources for passive neutron coincidence mass calibration. The reference sources were mounted in a zero-matrix SWB in the positions specified in the table.

Total Pu (g)	Source	Tube	Height (in)	Pu-240$_{eff}$ (g)
0.1	PDP1-0.1	J	15	0.006
0.5	PDP1-0.5	O	15	0.030
26.921	RANT25-1	O	15	1.85
76.95	RANT 25-1 RANT 50-1	O O	10 20	1.845 3.43
177.00	RANT 25-1 RANT 50-1 RANT 50-2 RANT 50-3	O O J P	10 20 20 15	1.845 3.43 3.43 3.43
190.003	RANT 25-1 RANT 50-1 RANT 50-2 RANT 50-3 PDP1-10 PDP1-3	O O J J P P	10 20 10 20 10 20	1.845 3.43 3.43 3.43 0.685 0.183
6.85	4WE*	O	15	13.58
17.4	4WE* MF-22*	O J	15 20	13.58 20.43

*Pu-238 sources that were decay corrected to March of 2011.

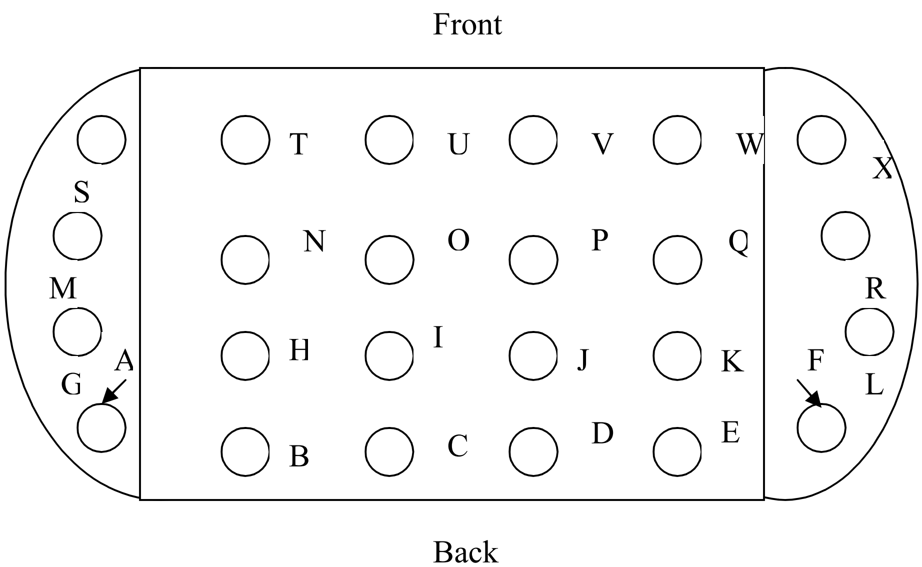

Figure 3: Top View, Cross Section of Zero Matrix Standard Waste Box Used for Calibration Measurements. This box is similar to the Performance Demonstration Program zero-matrix box.

The sources shown in Table 1 were then applied to create the calibration curve for the passive neutron coincidence rate in Figure 4.

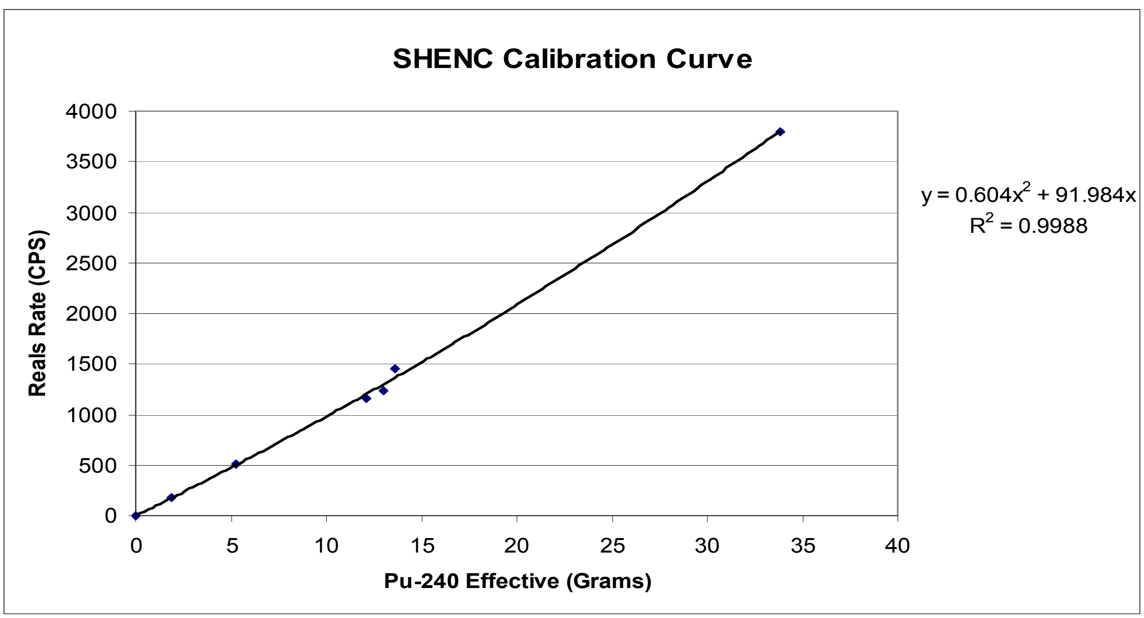

Figure 4. The calibration curve for the SHENC using the calibration loadings discussed in Table 1. This curve is used for Passive Neutron Coincidence counting measurements.

GAMMA CALIBRATION DESCRIPTION

The gamma ray efficiency calibration that relates count rates to decay rates was performed with a set of NIST-traceable gamma ray reference sources. These working reference materials were Eckert and Ziegler Catalog number EG-Line mixed gamma line reference sources, with a nominal combined activity of 120 μCi (60 μCi each of ^{241}Am and ^{152}Eu). The radioactive materials are dispersed in epoxy, and are encapsulated in aluminum tubes. The emitted gamma rays provide an effective calibration range from 59.5 keV to 1408 keV, suitable for measuring the radionuclides anticipated in LANL waste. These reference sources are traceable to the national standards database, and their certificates are reproduced in Appendix 1 of Reference [4]..

- Reference SWBs were assembled using a set of twelve gamma ray reference line sources in five different matrices and densities, including zero matrix (approximately 0.0 g/cm^3), Cardboard (0.152 g/cm^3), Cardboard/Plywood (0.297 g/cm^3), Cardboard/High Density Poly/Plywood (0.58 g/cm^3) and Plywood/Low Density Poly (0.788 g/cc). The reference SWBs have tubes positioned vertically in the matrix material, in a configuration such that the tubes are at different distances from the SWB axis. Figure 5 shows the tube configuration for the calibration SWBs that contain matrix material. The line sources extended along the full height of the SWBs and were loaded in a manner such that the response, upon rotation of the SWBs during the assay measurement, represented a uniform distribution of radioactive sources. The gamma ray reference sources are described in Table 2, and the locations are shown in Figure 5. The half-life for Eu-152 was taken from the Brookhaven National Nuclear Data Center (USDOE).

Table 2: Gamma calibration reference sources.

Source ID (S/N)	Nuclide	Half Life (y)	Activity (μCi)	Activity Ref. Date
H5-599	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.128± 3.48% 5.052 ± 3.50%	12/1/2010
H5-600	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.099 ± 3.48% 5.024 ± 3.50%	12/1/2010
H8-798	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.132± 3.48% 5.026 ± 3.50%	4/1/2011
H8-799	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.386 ± 3.48% 5.274 ± 3.50%	4/1/2011
H8-800	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.259 ± 3.48% 5.15 ± 3.50%	4/1/2011
H8-801	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.294 ± 3.48% 5.185 ± 3.50%	4/1/2011
H8-802	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.502 ± 3.48% 5.388 ± 3.50%	4/1/2011
H8-803	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.132 ± 3.48% 5.026 ± 3.50%	4/1/2011

H8-804	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.375 ± 3.48% 5.24 ± 3.50%	4/1/2011
H8-805	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.17 ± 3.48% 5.063 ± 3.50%	4/1/2011
H8-806	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.475 ± 3.48% 5.362 ± 3.50%	4/1/2011
H8-807	Am-241 Eu-152	432.7 ± 0.5 13.33 ± 0.04	5.175 ± 3.48% 5.068 ± 3.50%	4/1/2011

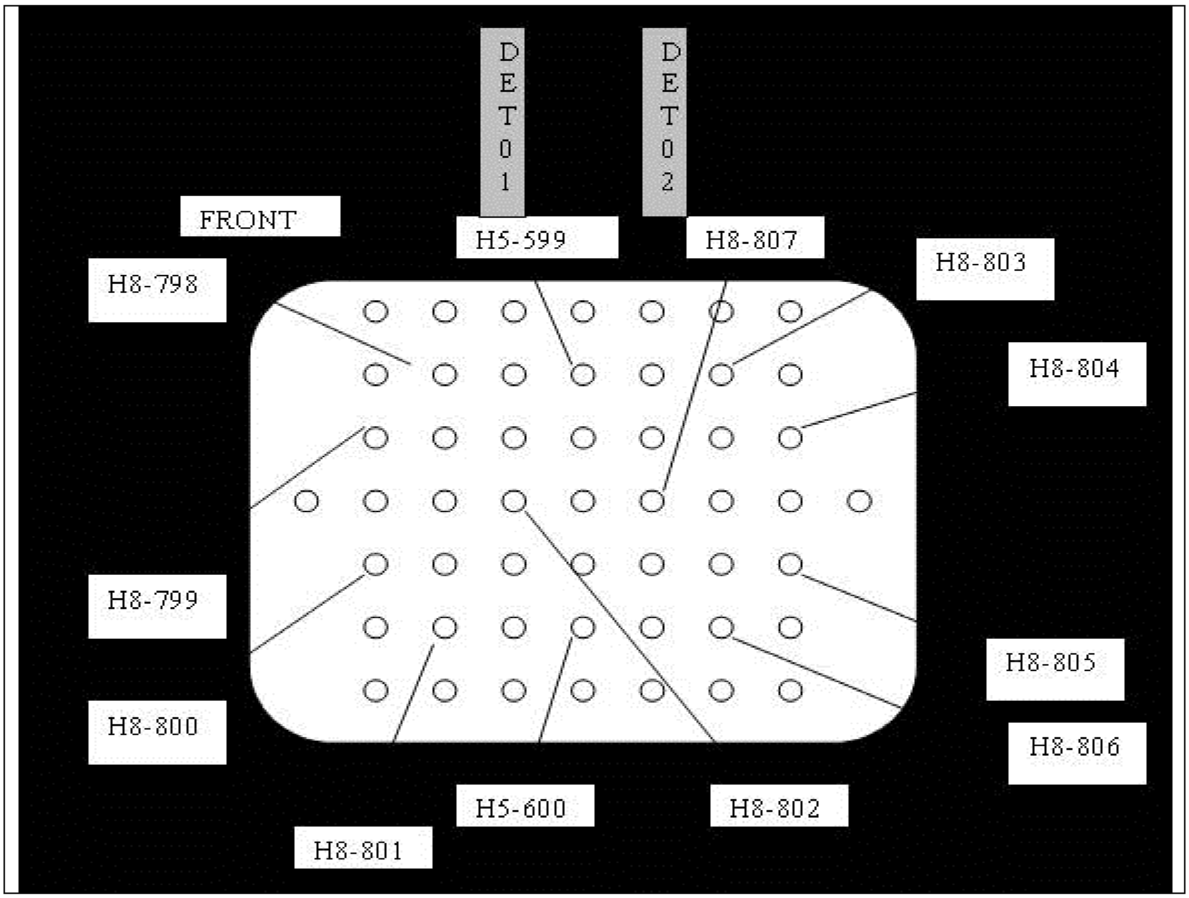

Figure 5: Line source distribution in the four calibration containers containing matrix material for the SHENC.

The sources listed in Table 2 were loaded into each matrix SWB as shown in Figure 5. Calibration assays were then performed for each matrix with varying count times to insure the proper amount of counts for each peak used in the calibration. Figure 6 shows the calibration curves generated from the data taken for the efficiency assays.

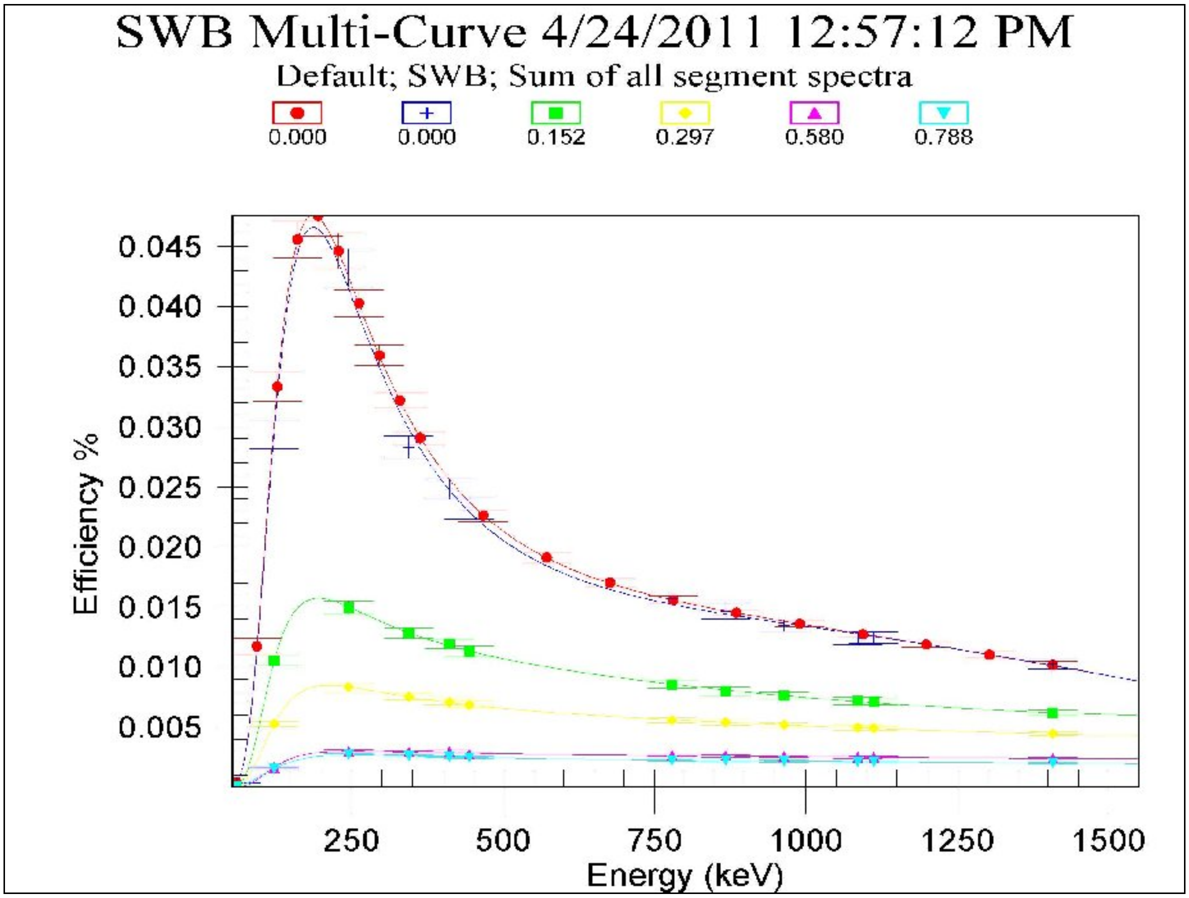

Figure 6: The above figure is the SHENC gamma density calibration curve using two High Purity Germanium Detectors. The above figure also contains a fit (red, filled circles) of an actual measurement at 0.000 g/cc.

During a gamma assay on the SHENC the SWB is rotated once during the assay. The SWB was also rotated in the same fashion during the calibration assays for the gamma modality of the SHENC. The field of view for each detector covers the complete area of the box. Since the system performs one rotation during an assay 4 different segments are acquired. These four segments are then averaged to produce the assay result for the gamma modality on the SHENC.

NEUTRON AND GAMMA CONFIRMATION RESULTS

All assay systems accepted into the WIPP certified program require confirmation measurements for each modality applied to a waste assay. The Transuranic Waste Acceptance Criteria in Table A-3.2 from document DOE/WIPP -02-03122 describes the requirements for system acceptance. The upper limit for the %RSD (precision) values for six measurements is 14%. The confirmation measurement upper limit for R (recovery) is ± 30%. The results for the confirmation measurements are presented in Table 3. All gamma results passed both precision and accuracy requirements quoted in the Transuranic Waste Acceptance Criteria Table A-3.2 [5].

Table 3: Gamma source confirmation loadings with accuracy (%R) and precision (%RSD) results.

Standard ID	Pu Mass (g)	Standard Matrix	Position of Standards in SWB	Simulated or Actual Waste Matrix	# of Assays	Gamma Results Accuracy and Precision
NTP-0001 **Total**	0.1316 **0.1316**	DE[#]	Tube O Ht: 15”	Zero Matrix SWB	6	%R=85.5 %RSD=7.73
RANT25-1 **Total**	26.88 **26.88**	DE	Tube O, Ht: 15”	Zero Matrix SWB	6	%R=93% %RSD=6.53
NTP-0164 NTP-0156 **Total**	64.89 49.82 **114.7**	DE	Tube O, Ht: 15” Tube J, Ht: 15”	Zero Matrix SWB	6	%R=93.3 %RSD=4.96
NTP-0164 NTP-0156 NTP-0148 PDP40.0Pu1 **Total**	64.89 49.89 30.00 40.00 **184.8**	DE	Tube O, Ht: 15” Tube J, Ht: 15” Tube I, Ht:15” Tube P, Ht: 15”	Zero Matrix SWB	6	%R=86.9 %RSD=6.09
NTP-0164 NTP-0156 NTP-0148 NTP-0140 NTP-0007 NTP-0199 PDP40.0Pu1 PDP40.0Pu2 **Total**	64.89 49.89 30.00 15.025 7.51 26.36 40.01 40.04 **272.70**	DE	Tube J, Ht: 10” Tube J, Ht: 20” Tube P, Ht: 10” Tube P, Ht: 20” Tube O, Ht: 10” Tube I, Ht: 20” Tube O, Ht: 20” Tube I, Ht: 10”	Zero Matrix SWB	6	%R=91.9 %RSD=2.75
MF40F* (Pu-238)	**7.0**	DE	Tube O, Ht: 15”	Zero Matrix SWB	6	%R=123.4 %RSD=10.52
MF-76* MF-40F* (Pu-238) **Total**	10.07 7.0 **17.07**	DE	Tube O, Ht: 15” Tube J, Ht: 15”	Zero Matrix SWB	6	%R=108.3 %RSD=12.04

*Pu-238 sources that were decay corrected to March of 2011.
[#]DE = Diatomaceous Earth

Table 4: Neutron source confirmation loadings with accuracy (%R) and precision (%RSD) results. *Pu-238 sources that were decay corrected to March of 2011. #DE = Diatomaceous Earth.

Standard ID	Pu Mass (g)	Standard Matrix	Position of Standards in SWB	Simulated or Actual Waste Matrix	# of Assays	Neutron Results Accuracy and Precision
NTP-0001 **Total**	0.1316 **0.1316**	DE#	Tube J, Ht: 15”	Zero Matrix SWB	6	%R=121.0 %RSD=22.0
NTP-0085 NTP-0092 **Total**	0.3062 0.3052 **0.6114**	DE	Tube O, Ht: 15” Tube J, Ht: 15”	Zero Matrix SWB	6	%R=95.2 %RSD=6.82
NTP-0148 **Total**	30.0 **30.0**	DE	Tube O, Ht: 15”	Zero Matrix SWB	6	%R=96.2 %RSD=0.29

Standard ID	Pu Mass (g)	Standard Matrix	Position of Standards in SWB	Simulated or Actual Waste Matrix	# of Assays	Neutron Results Accuracy and Precision
NTP-0164 NTP-0156 **Total**	64.89 49.89 **114.8**	DE	Tube O, Ht: 10” Tube O, Ht: 20”	Zero Matrix SWB	6	%R=99.1 %RSD=0.52
NTP-0164 NTP-0156 NTP-0148 **Total**	64.885 49.89 30.0 **144.77**	DE	Tube J, Ht: 10” Tube O, Ht: 20” Tube P, Ht: 15”	Zero Matrix SBW	6	%R=98.8 %RSD=1.00
NTP-0164 NTP-0156 NTP-0148 NTP-0140 NTP-0199 NTP-0007 **Total**	64.89 49.89 29.99 15.025 25.36 7.51 **192.7**	DE	Tube O, Ht: 10” Tube O, Ht: 20” Tube J, Ht: 10” Tube J, Ht: 20” Tube P, Ht: 10” Tube P, Ht: 20”	Zero Matrix SWB	6	%R=97.7 %RSD=0.46
NTP-0164 NTP-0156 NTP-0148 NTP-0140 NTP-0007 NTP-0199 PDP40.0Pu1 PDP40.0Pu2 **Total**	64.89 49.89 30.0 15.025 7.51 26.36 40.01 40.04 **272.70**	DE	Tube O, Ht: 10” Tube O, Ht: 20” Tube J, Ht: 10” Tube J, Ht: 20” Tube P, Ht: 10” Tube I, Ht: 20” Tube P, Ht: 20” Tube I, Ht: 10”	Zero Matrix SWB	6	%R=95.4 %RSD=0.32
MF40F* (Pu-238)	**7.0**	DE	Tube O, Ht: 15”	Zero Matrix SWB	6	%R=106.7 %RSD=0.30
MF-40F* MF-76* (Pu-238) **Total**	7.0 10.07 **17.07**	DE	Tube O, Ht: 15” Tube J, Ht: 20”	Zero Matrix SWB	6	%R=98.0 %RSD=0.20

The neutron confirmation results for the SHENC met the WAC requirements in Reference [5] except for the lowest mass loading. The 0.13 gram mass loading met the accuracy requirements for the WAC however failed the precision requirements. The system applies only the gamma modality when performing assays in the 0.13 g Pu mass range. This is an administrative control performed by the Expert Analyst.

ISOTOPIC MEASUREMENTS

The Isotopic Measurements performed on the SHENC use either MGA version 9.3H or FRAM version PC 4.4. Each assay is evaluated by the Expert Analyst for isotopic content. MGA, FRAM or Acceptable Knowledge isotopics are then applied to the assay depending upon the evaluation.

DISCUSSION

The SHENC was successfully upgraded to efficiently measure the complex waste streams at Los Alamos National Laboratory. A new PLC was successfully added to the system for Add-A-Source control. A new shift register was added to the SHENC (JSR-15) which provides more flexibility if replacement is required. The addition of the density based calibration on the gamma modality of the system increases the effectiveness of the SHENC for high alpha-n producing waste. Upgrading the system to new software also helped to standardize the program at Los Alamos. Only one new procedure was initiated for the integration of the SHENC. Add-A-Source measurements were effectively integrated with the system based on the confirmation results and positive results during two Department of Energy Performance Demonstration Program Box Cycles [6]. Total measurement uncertainty estimates were completed for both the gamma modality and the neutron modality of the system.

The SHENC was reviewed by both the Department of Energy and the Environmental Protection Agency in 2011. The certification letter for the SHENC was issued on November 16, 2011 [7]. With the certification letter issued to the Central Characterization Project the SHENC can perform waste measurements on SWB’s at Los Alamos for shipment to WIPP.

REFERENCES

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