Calibration of a Passive Neutron Counter with ^{242}Pu Standards

16450

Joseph Wachter[1], jwachter@canberra.com, 505-470-2567
Adam Gallegos[2], agallegos@pscnda.com, 505-670-4129
Robert McElroy[3], mcelroyrd@ornl.gov, 865-576-2107
Marcel Villani[1], marcel.villani@canberra.com, 203-639-2370
Bruce Gillespie[1], bruce.gillespie@canberra.com, 509-539-0760
Sean Stanfield[1], sean.stanfield@canberra.com, 505-231-0557
Joe Harvill[4], joe.harvill@wipp.ws, 575-302-8647

1. Canberra Industries, 1350 Central Ave, Suite 300, Los Alamos, NM 86544
2. Pajarito Scientific Corp, 1350 Central Ave, Suite 300, Los Alamos, NM 86544
3. Oak Ridge National Laboratory, Oak Ridge, TN
4. Central Characterization Program, 4021 National Parks Hwy, Carlsbad, NM 88220

At the Los Alamos National Laboratory TA 55 facility, a large quantity of Heat Source (HS) and MT42 (primarily ^{242}Pu) has been designated for disposal as TRU waste. The waste, to be packaged in either 55 gal drums or Pipe Overpack Containers (POCs), is scheduled to be assayed with the TA 55 HENC 3 system. The system is owned and operated by the Nuclear Waste Partnership (NWP) under a contract with the Carlsbad DoE field office. The HENC 3 is a combined passive neutron/gamma assay system. The neutron component of the system was previously calibrated up to 21.9 g of ^{240}Pu effective. This corresponds to approximately 9 g of typical HS plutonium and 13 g of MT42 waste. In order to reduce packaging efforts and radiation dose for waste handling personnel, LANL has requested an increase in the HENC 3 calibration limit to approximate the maximum allowable shipping and storage limits for TRU waste.

To increase the limits, NWP has obtained the use of well characterized MT42 source materials. The sources, developed and owned by LANL, range from to 5 g to 200 g of plutonium enriched to 86.6% ^{242}Pu. Using these sources in combination, multiple measurements were performed with both 55-gal drums and POCs to determine their suitability for extending the neutron calibration range of the system. The results of the measurements were (1) compared to the existing calibration for consistency and (2) used to extend the calibration range to 307 g of ^{240}Pu effective.

This report will discuss the elements of the neutron component of the HENC 3 system, the calibration extension effort, problems encountered, and the results of

the measurements. As a result of the extension, HS and MT42 materials can be assayed in 55-gal drums and POCs up to their allowable shipping limits.

INTRODUCTION

Contact-handled transuranic waste with plutonium enriched in ^{238}Pu (Heat Source-HS plutonium) and ^{242}Pu (MT42 plutonium) must be removed from the Plutonium Facility at Los Alamos National Laboratory (LANL). In order to dispose of the waste with minimal handling, it will be packaged such that the radioactivity in each waste container is close to the maximum amount allowed by shipping and storage regulations for the WIPP disposal site[1,2]. The activity is also used to calculate other regulated quantities relating to decay heat, criticality risk, dose, and transportation hazard. At present, the waste is to be packaged in approved 55-gal drums and Pipe Overpack Containers (POC) prior to assay. Shipments in Criticality Control Overpack (CCO) containers may be used in the future for this waste. For the purposes of this discussion, the most important quantities and their limits are as follows:

Quantity	55-gal Drum	POC
Fissile Gram Equivalent (FGE)	≤ 200 g	≤ 200 g
Plutonium Equivalent Curies (PE-Ci)	≤ 80 PE-Ci	≤ 1800 Pe-Ci

The HENC 3 nondestructive assay (NDA) system, located at the Plutonium Facility at LANL, is a combined gamma and neutron assay instrument. It was previously designated and approved for transuranic waste characterization of weapons grade plutonium[3] and small amounts of Heat Source and MT 42 plutonium. Because of the system's proximity to the facility where the HS and MT42 waste is stored, it will be used to assay these materials. However, its previous calibration for weapons grade plutonium did not extend to the anticipated packaging limits needed to minimize waste handling. For this reason, an effort was undertaken to extend the operating limits for the neutron component of the system to enable it to assay larger quantities of plutonium enriched in ^{238}Pu and ^{242}Pu. The extension was completed using a set of MT42 standards containing between 5.1 and 194 grams of plutonium enriched to 86.6% of ^{242}Pu. This correlates to between 8.0 and 307 g of ^{240}Pu *effective* (^{240}Pu *eff)*, the term of merit used for most passive neutron coincidence counters.

In order to certify the instrument for measurements of larger quantities of HS and MT42 transuranic waste, three tasks were undertaken. First a comparison of the original neutron calibration confirmation results using Weapons Grade (WG) plutonium with recent measurements of MT42 sources was performed in order to

establish consistency with the original calibration. Next, the use of linear calibration parameters for non multiplying forms of plutonium was demonstrated. Finally, an extension of the neutron calibration and operating range of the HENC 3 for non multiplying forms of plutonium was established.

This report will discuss the HENC 3 system and the effort to extend the calibration range to accommodate larger quantities of enriched ^{238}Pu and ^{242}Pu waste. A description of the system and the methodology used for the calibration extension will first be presented. Following that, the sources that were used to confirm the extended calibration will be described. Finally, the results of those confirmation measurements and the new calibration range will be offered.

LANL HENC 3 SYSTEM

The HENC 3 (Figure 1) system is a combined neutron and gamma assay system that is used to assay TRU waste generated at the TA 55 facility at LANL. It consists of a 30% efficient neutron counting module, JSR-14 neutron coincidence signal processor, and an Add-a-Source component for matrix correction. A single high-resolution gamma detector is used to quantify the activity of individual isotopes in the waste and determine the relative isotopic composition of the plutonium and ^{241}Am isotopes. A low energy photon filter is placed in front of the gamma detector face which is 51 cm from the axial center line of the turntable. The system is used to assay both debris and homogeneous waste with densities between 0.01 g/cc and 2.5 g/cc. Separate calibrations were performed for three approved TRU waste container types: 55-gal drum, POC, and CCO. The original calibration models were based on container designs and site knowledge of expected waste matrix materials and were determined without the use of radioactive sources. Confirmation was completed by performing multiple measurements over the expected activity range in a non interferring matrix to verify that the modeling had been correctly performed and installed. The original source loadings ranged from less than 1 g of Pu to 315 g and were performed for the three container types using WG sources. Afterwards, calibration validation measurements were performed with plutonium sources in surrogate matrix drums containing combustibles (ρ = 0.16 g/cm^3), metals (ρ = 0.26 g/cm^3), glass (0.54 g/cc), and cement (ρ = 1.85 g/cm^3) matrices. These measurements tested the validity of the modeled efficiency parameters over a range of densities. The original confirmation and validation measurements verified that the calibrations were appropriate for the container types, matrices, and plutonium mass range. Prior to acceptance, the total measurement uncertainty

Figure 1. HENC 3 system.

budget was also estimated. Determination of the budget was performed using measurements, modeled estimates of the variability due to source and matrix heterogeneity, and estimates based on experience. It included uncertainty factors such as background, statistical uncertainty in the quantitative and Pu isotopic measurements, uncertainty associated with calibration, source and matrix heterogeneity effects, uncertainty due to detector positioning, and other potential sources of measurement uncertainty. The system was installed, tested, and certified to assay TRU waste during 2014[3].

For the calibration range extension described in this report, only the neutron component was re-evaluated. No other changes to the operation of the HENC 3, including gamma, Add-A-Source, QC, or calibration measurement parameters were validated.

DESCRIPTION OF ^{242}PU STANDARDS

The $^{242}PuO_2$ standards used for the calibration extension were originally packaged in 4" diam x 5" cans with welded lids. They ranged in ^{240}Pu *effective* content from 8 g to 307 g. One of the standards contained approximately 700 g of KCl + NaCl diluent whereas the others contained no diluent. All of the cans were later overpacked into 6.8" diam x 9.1" tall stainless steel outer cans because of deterioration due to an acidic environment while they were inside processing gloveboxes. The standards were fabricated at Los Alamos National Laboratory during 1990 – 1991. In 2014, they received multiple calorimetry and plutonium isotopic measurements to confirm their original plutonium isotopic mass values in an effort to certify them to the national standards database.

In passive neutron counters such as the HENC 3, spontaneous fission neutrons are thermalized before being detected by ^{3}He proportional counters. In this process, all energy information from the neutrons is lost during thermalization. Moreover, the energy, spontaneous fission multiplicity, and neutron multiplicity moments for ^{238}Pu, ^{240}Pu, and ^{242}Pu are nearly identical. For these reasons, the detection of a neutron from ^{242}Pu looks exactly like a neutron from either ^{238}Pu or ^{240}Pu. So neutron calibration or confirmation of the HENC 3 can be performed with standards from either ^{238}Pu, ^{240}Pu, or ^{242}Pu provided corrections are made for multiplication effects and their relative spontaneous fission yields. The correction for spontaneous fission yields is provided by the normalizing term ^{240}Pu effective (^{240}Pu *eff*) mass where

$$^{240}\text{Pu } eff = 2.52\ ^{238}\text{Pu} + {}^{240}\text{Pu} + 1.69\ ^{242}\text{Pu}$$

where ^{238}Pu, ^{240}Pu, and ^{242}Pu refer to the masses of the respective isotopes

This claim was validated by comparing the original neutron calibration confirmation measurements using Weapons Grade (WG) sources with measurements using MT42 standards with ^{242}Pu as the majority isotope. The measurements were performed for both 55-gal drums and POCs.

COMPARISON OF MT42 AND WEAPONS GRADE MEASUREMENTS

To confirm that measurements of ^{242}Pu materials yield the same results as those from MT52 sources, the results of the MT42 confirmation measurements were compared to those performed during the original WG confirmation where the quantities of ^{240}Pu *eff* overlap in the two sets of measurements. During the original confirmation measurements, the quantity of ^{240}Pu *eff* ranged between 0.030 g and 21.83 g for both 55-gal drums and POCs. During recent confirmation measurements of the MT42 sources, quantities of 8.00 g ^{240}Pu *eff* and 14.8 g ^{240}Pu *eff*, were completed In Figures 1a and 1b below, the results of the two sets of measurements are compared for 55-gal drums and 12" diam POCs. Comparisons are provided only for coincidence mode analysis as the multiplicity mode results were not described in the original calibration report. Percent Recovery (%R) for the comparative measurements refers to the ratio of measured values to the nominal ^{240}Pu *eff* values of the standards expressed as a percent. The reported results for both MT42 and WG sources are the averages of six confirmation measurements of each source.

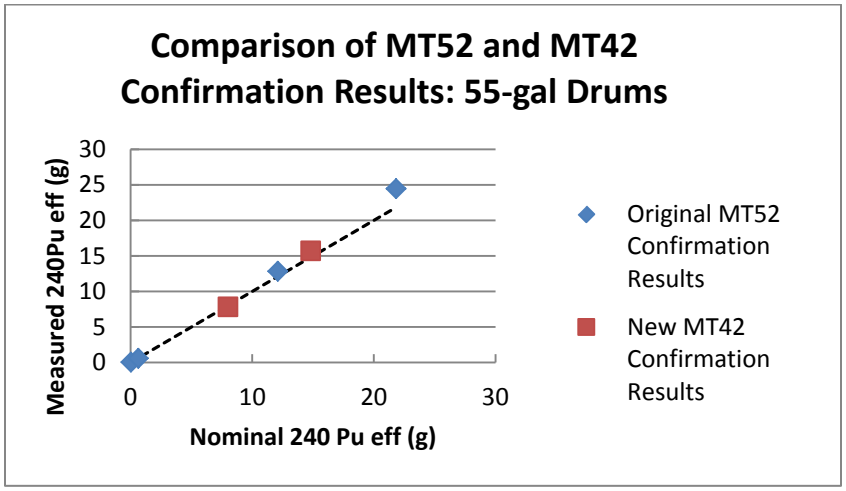

Figure 1a. Comparison of original MT52 confirmation results with recent MT42 confirmation measurements for 55-gal drums.

For both 55-gal drums and 12" POCs, the results indicate that the sources enriched in ^{242}Pu provide roughly the same %R as the WG sources which were used to confirm the calibration in the original calibration report. This validates that

measurements of ^{242}Pu materials yield the same results as those from MT52 sources when there is less than 21.83 g of ^{240}Pu *eff*.

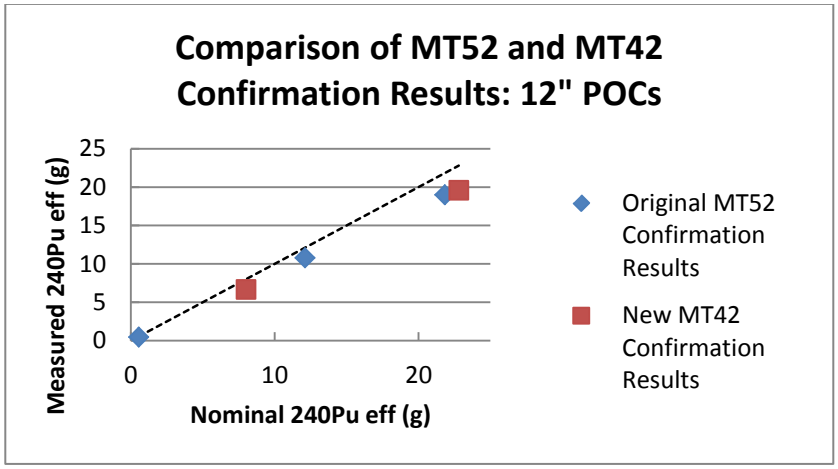

Figure 1b. Comparison of original MT52 confirmation results with recent MT42 confirmation measurements for 12" POC containers.

CALIBRATION CONFIRMATION FOR NON MULTIPLYING FORMS OF PLUTONIUM

When the HENC 3 system was originally calibrated through the use of known physical constants and measured physical parameters, a quadratic formulation was used for the coincidence mode calibration:

$$R = a_0 + a_1 m_{240} + a_2 (m_{240})^2 \quad (1)$$

where R is the measured Reals rate corrected for background and dead time effects

a_0 is the intercept of the calibration curve. This term is 0 for the HENC 3.

a_1 is the coefficient for the linear term. This is 53.1 -/g for the HENC 3.

a_2 is the coefficient for the quadratic term. This is 0.5194 -/g^2 for the HENC 3.

m_{240} is the mass of ^{240}Pu *eff* .

In the NDA2000 software, this form is referred to as the Reals formulation. These parameters work very well for multiplying forms of plutonium (for example, WG waste) and for relatively small amounts of non multiplying forms of plutonium (for example, MT42 and Heat Source plutonium). But for larger amounts of non multiplying plutonium waste, an alternative, and linear, calibration must be used. The alternative calibration is also derived from known physical constants and measured parameters, and has the form:

$$R = a m_{240} \quad (2)$$

where a = 54.829 for the HENC 3

In the NDA2000 software, this formulation of the calibration is referred to as the Multiplication Corrected Reals (MC Reals).

When measuring plutonium waste, the original form of the calibration (Equation 1) remains in use for multiplying plutonium samples and the calibration range will remain the same as stated in earlier versions of this calibration report (0.030 to 21.9 g of ^{240}Pu *eff*). However, for future measurements of non multiplying plutonium such as Heat Source and MT42, the linear formulation (Equation 2) will be used.

In order to determine that a new calibration of an NDA instrument is correctly established, DOE/WIPP-02-3122 Appendix A[2] requires that the accuracy and precision of the instrument be confirmed by performing replicate measurements of reference sources in non-interfering matrices.

The neutron calibration was confirmed by performing six measurements of MT42 plutonium sources[4] in both an empty 55-gal drum and an empty POC. For 55-gal drums, the plutonium mass ranged from 8.00 to 307 g of ^{240}Pu *eff*. POCs were confirmed with sources ranging from 8.00 to 461 g of ^{240}Pu *eff*. Data for both the coincidence results using Equation (2) and multiplicity results were collected. Figures 2a and 2b present the results of the confirmation measurements graphically for 55-gal drums and POCs, respectively.

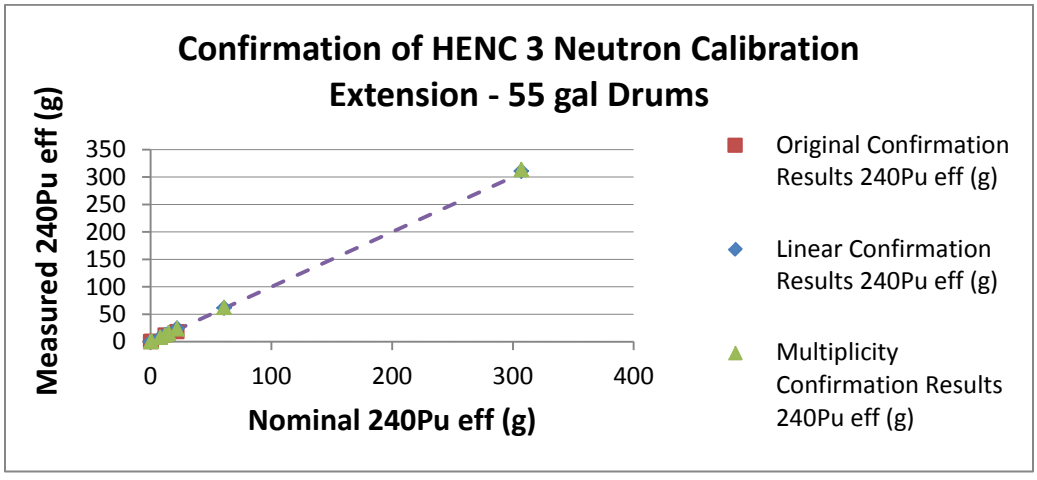

Figure 2a. Linear and Multiplicity confirmation results for 55-gal drums. The dotted line indicates perfect agreement between the measurements and ^{240}Pu *eff* values for the sources.

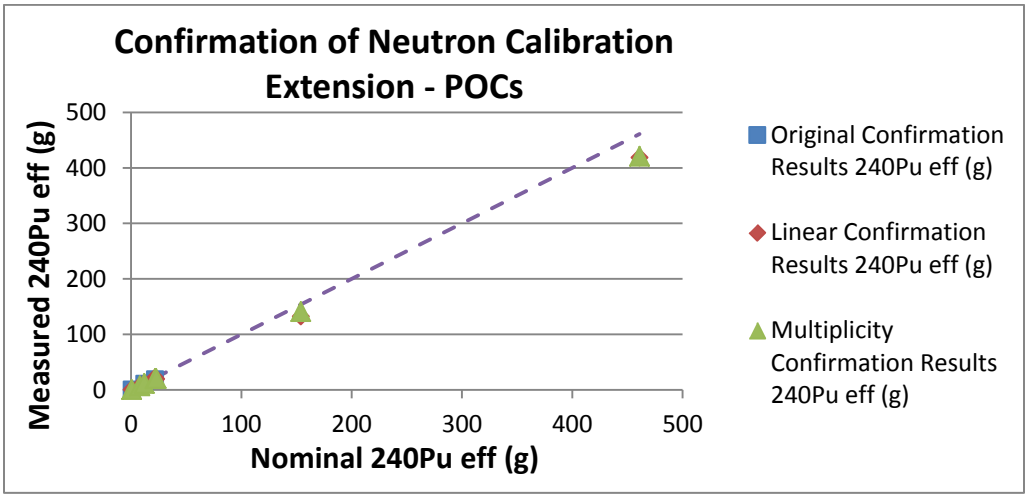

Figure 2b. Linear and Multiplicity confirmation results for POC containers. The dotted line indicates perfect agreement between the measurements and ^{240}Pu *eff* values for the sources.

The confirmation measurement results were evaluated against the criteria specified in DOE/WIPP-02-3122[2] Appendix A, Table A-2[2]. According to that document, 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 confirmation measurements performed.

Using the results provided above, the following neutron operating ranges for HENC 3 neutron measurements of TA 55 TRU waste are indicated:

- Multiplying forms of plutonium, including Weapons Grade Pu in 55-gal drums and POCs: 0.030 to 21.83 g of ^{240}Pu *eff*. This limit remains unchanged. These measurements are to be performed using the quadratic, or Reals, calibration formulation (Equation 1).
- Heat Source and MT42 Pu in 55-gal drums: LLD to 307 g of ^{240}Pu *eff*. These measurements are to be performed using the linear, or Multiplication Corrected Reals (MC Reals) calibration formulation (Equation 2).
- Heat Source and MT42 Pu in POCs: LLD to 461 g of ^{240}Pu *eff*. These measurements are to be performed using the linear, or Multiplication Corrected Reals (MC Reals) calibration formulation (Equation 2).
- Both multiplicity and MC Reals coincidence modes may be used in the operating range for non multiplying forms of plutonium.

- Mixtures of multiplying and non multiplying plutonium waste: TRU waste packagers will take every effort to separate multiplying and non multiplying plutonium waste. In cases where they are comingled, the measured results are acceptable using the MC Reals (Equation 1) formulation provided the total mass of ^{240}Pu *eff* is $\leq$ 21.8 g. This corresponds to the upper limit of the operating range determined during the original calibration.

CONCLUSIONS

This report has shown that (1) MT42 standards may be used for calibration of passive neutron coincidence counters, (2) a linear formulation of the calibration term is appropriate for large amounts of non multiplying plutonium, and (3) the neutron calibration can be extended to, at least, 461 g of ^{240}Pu *eff*. The extended capability of the HENC 3 can now be used to assay quantities that more closely approach the maximum allowable shipping and storage limits for TRU waste.

REFERENCES

1. TRU Waste Authorized Methods for Payload Control (CH-TRAMPAC Document), Rev 4, April 2014.
2. Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant, DOE/WIPP-02-3122, Ver 7.4, April 2014.
3. J. Wachter et al, Model-Based Approaches to Calibration and Certification of Nondestructive Assay Systems, *Proc of WM2015 Symposia*, Phoenix, 2015.
4. STDB242C1, STDB242C2, STDB242C3, STDB242C4,STDB242C7 and STDB242C8 Standards Requalification, W. Salazar, Los Alamos National Laboratory Report *LA-UR 15-27985*, Los Alamos, NM, May 11, 2015.