Innovations in the Assay of Un-Segregated Muti-Isotopic Grade TRU Waste Boxes with SuperHENC and FRAM Technology

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ABSTRACT

The Super High Efficiency Neutron Coincidence Counter (SuperHENC) was originally developed by BIL Solutions Inc., Los Alamos National Laboratory (LANL) and Rocky Flats Environmental Technology Site (RFETS) for assay of transuranic (TRU) waste in Standard Waste Boxes (SWB) at Rocky Flats. This mobile system was a key component in the shipment of over 4,000 SWBs to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico. The system was WIPP certified in 2001 and operated at the site for four years. The success of this system, a passive neutron coincidence counter combined with high resolution gamma spectroscopy, led to the order of two new units, delivered to Hanford in 2004. Several new challenges were faced at Hanford: For example, the original RFETS system was calibrated for segregated waste streams such that metals, plastics, wet combustibles and dry combustibles were separated by “Item Description Codes” prior to assay. Furthermore, the RFETS mission of handling only weapons grade plutonium, enabled the original SuperHENC to benefit from the use of known Pu isotopics. Operations at Hanford, as with most other DOE sites, generate un-segregated waste streams, with a wide diversity of Pu isotopics. Consequently, the new SuperHENCs are required to deal with new technical challenges. The neutron system’s software and calibration methodology have been modified to encompass these new requirements. In addition, PC-FRAM software has been added to the gamma system, providing a robust isotopic measurement capability. Finally a new software package has been developed that integrates the neutron and gamma data to provide a final assay results and analysis report. The new system’s performance has been rigorously tested and validated against WIPP quality requirements. These modifications, together with the mobile platform, make the new SuperHENC far more versatile in handling diverse waste streams and allow for rapid redeployment around the DOE complex.

INTRODUCTION

The SuperHENC performs nondestructive assay (NDA) in order to determine radionuclide contents of drums and standard waste boxes up to a maximum envelope of 138.4 cm (54.5 inches) wide by 94.0 cm (37 inches) high by 180.3 cm (71 inches) long. The system combines a high efficiency neutron assay chamber with a high resolution gamma spectroscopy system in a single transportable trailer.

At Hanford, a new SuperHENC system (SHENCA) is being used the Waste Receiving and Processing (WRAP) facility to measure TRU heterogeneous debris waste with a required lower limit of detection of 100 nCi/g for sentencing to the Waste Isolation Pilot Plant (WIPP) [1, 2]. This system is shown in Figure 1. A second identical system (SHENCB) is currently deployed at the Plutonium Finishing Plant (PFP) at Hanford in support of the site’s Materials Control and Accountability (MC&A) program.
The neutron assay chamber utilizes a six sided arrangement of polyethylene moderated He-3 detectors. The detectors are filled to ten atmospheres pressure and have various active lengths. The exterior of the neutron chamber is clad with eight inches of polyethylene to shield against exterior neutron sources. Passive neutron coincidence counting and multiplicity techniques [3, 4] are used to quantify the Pu-240 effective (Pu-240e) mass content of the waste container. Matrix correction is achieved by use of the Add-A-Source (AAS) technique. This method uses a Cf-252 source that is automatically transferred to reference positions on the floor of the assay chamber by a Teleflex cable. When not in use, the source is stored in a polyethylene pig.

The SuperHENC Gamma Energy Analysis System (SGEAS) comprises a single 30mm thick coaxial High Purity Germanium (HPGe) detector viewing SWBs placed on a turntable. A filter of 1.09mm cadmium is located in front of the HPGe detector (within a shield/collimator) in order to reduce the strong gamma emission from Am-241 60keV gamma rays. An optional additional 10mm steel filter is available for assay of high gamma activity waste containers. The SGEAS is used for two types of measurement:

- Relative ratio mode: Isotopic ratios for Pu and other isotopes of interest are determined by analyzing the acquired spectra with the PC-FRAM isotopic code. Default isotopic ratios are used where counting statistics are poor.
• Absolute mode: Some radionuclides may be directly determined from analysis of the gamma spectrum. For most TRU nuclides, the low energy associated with the gamma emission combined with the attenuating properties of the matrix in the SWB usually results in a large total measurement uncertainty (TMU) for the absolute gamma mode. The ratio method is usually preferred because the neutron assay is more accurate. However, in some circumstances absolute gamma assay can be used for direct radionuclide quantification. Examples include waste for which the neutron coincidence signal is subject to significant interference from high (alpha,n) emission or where uranium isotopes are present in the absence of plutonium.

SOFTWARE IMPROVEMENTS

The SuperHENC utilizes a custom tailored version of the LANL Neutron Coincidence Counting (NCC) software [5] (SUPRHENC.exe Version 2.0). The software controls data collection, analysis and the AAS movement, and contains all exception handling functions. Modifications were preformed to allow for interface to updated machine control hardware. Additionally, the modified machine interface software now resides in a Dynamic Link Library (DLL). This facilitates future upgrades, without the need for modification of the main SuperHENC software.

The Gamma HPGe sub-system can be equipped with any data acquisition package which is capable of exporting PC-FRAM readable raw data files. This allows for the greatest flexibility to plant operations. Whether the plant has a preferred off-the-shelf acquisition platform (such as Ortec’s Maestro-32 or Canberra’s Genie 2000), or custom BIL Solutions, Inc. acquisition packages, the system is designed to easily fit into established site operating procedures. For the Hanford SuperHENCs’, Maestro-32 is currently used to control an Ortec DSPEC jr 2.0 multi-channel analyzer.

PC-FRAM [6] is utilized for advanced Gamma Isotopic Analysis of the HPGe acquired data. The use of PC-FRAM for analysis grants the system the flexibility of using any number of data acquisition systems based upon operational preference.

BIL Solutions, Inc. has created a Neutron Gamma Integration (NGI) package in Visual C++. This software provides for the integration of data from the SuperHENC Neutron and Gamma Subsystems. The NGI software outputs a WIPP compliant final report. The NGI software has been created in a modular (DLL) based fashion which allows for easily customizable options to meet specific plant needs. For example, should a plant require interface to an existing Waste Management Database, modification can be easily made to the Data Archiving DLL to send pertinent fields to such database.

CALIBRATION

The SHENCA neutron system was initialized with coincidence counting electronics parameters shown in Table I. These settings were optimized during factory testing. An Ortec Advanced Multiplicity Shift Register (AMSR-150) is used for the coincidence analysis.

Table I. SHENCA Coincidence Counting Parameters for AMSR-150.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Setting</th>
</tr>
</thead>
<tbody>
<tr>
<td>Predelay (µs)</td>
<td>1.5</td>
</tr>
<tr>
<td>Gate length (µs)</td>
<td>128</td>
</tr>
<tr>
<td>High Voltage</td>
<td>1720</td>
</tr>
<tr>
<td>Die Away Time (µs)</td>
<td>55</td>
</tr>
</tbody>
</table>
The calibration comprises several steps. These elements include 1) mapping chamber efficiency with a neutron source, 2) setting up the add-a-source, 3) constructing a Monte Carlo model for the system, 4) obtaining calibration measurements, 5) establishing the coincidence calibration curve, 6) establishing multiplicity calibration parameters and 7) calibration confirmation using independent plutonium standards. Each of these steps is discussed in more detail below.

**Chamber Efficiency**

Chamber efficiency is defined as the sum of all neutron totals (singles) channel count rates divided by source neutron emission rate. A Cf-252 source was positioned at 96 different coordinates within the SHENCA neutron cavity. A 3-dimensional plot of chamber efficiency is shown in Figure 2. The x- and y-axes indicate the location of the source in the horizontal plane of the SWB chamber based on an arbitrary reference system. The z-axis indicates efficiency (note that the scale is from 37.0% to 39.4%). The chamber efficiency averaged over all 96 positions was determined to be (38.23 +/- 2.22) %.

![Chamber Efficiency Maps](image)

**Fig. 2.** Hanford SHENCA chamber efficiency map - % singles (totals) efficiency
**Add-A-Source (AAS) Set Up**

The AAS is a Cf-252 source that travels through a Teleflex cable and stops at pre-selected positions on the floor of the assay chamber. The AAS is used for two functions: (i) Matrix Correction – the software calculates the measured response to the AAS, compares this to a reference count and calculates the matrix correction factor, (ii) Normalization – this is a simple and quick check on the empty neutron chamber counting efficiency compared to a reference initial source measurement (used for measurement control).

For SWB matrix correction measurements, six AAS positions were selected such that their spatial relationship in the horizontal plane of the chamber floor approximated a uniform area sample over the SWB footprint. Reference count rates were determined at each AAS position.

**Monte-Carlo Model**

A Monte Carlo N-Particle (MCNP) [7] model was used to determine the relationship between AAS response and matrix correction factor. Various waste compositions and densities were modeled for the system [8]. The AAS matrix correction factor \( (CF) \) is calculated from the following expression:

\[
CF = 1 + a + b \Delta + c \Delta^2 + d\Delta^3 \quad \text{(Eq. 1)}
\]

where \( a, b, c, \) and \( d \) are calibration coefficients determined from MCNP. For SWBs these coefficients have been determined to be \( a = -5.1366 \times 10^{-2}, b = 2.7148 \times 10^{-1}, c = 2.566 \times 10^0 \) and \( d = -5.201 \times 10^{-1} \).

\[
\Delta = r - 1 \quad \text{(Eq. 2)}
\]

where \( r \) is the ratio of the (decay corrected) add-a-source reference doubles rate (i.e., with empty box) to the measured add-a-source doubles rate (i.e., with real waste box). The reference and measured doubles rates are the average of the six AAS positions.

This correction factor is then applied to the doubles rate (measured with a waste box in the chamber and the add-a-source retracted) to return the corrected “empty box” doubles rate which is in turn used to determine Pu-240e mass (the Pu-240e mass calibration curve is determined with an empty box).

It is important to understand that the MCNP modeling is a one-time process performed before or during calibration. The AAS calibration coefficients are input into the software as part of the calibration process. In measurements of real waste, matrix correction is performed real-time and no prior knowledge of matrix category is required. Consequently, the operator is not required to input matrix composition.

The efficacy of the MCNP derived matrix correction is verified by a set of calibration confirmation measurements described later.

**Calibration Measurements**

Calibration was performed with an empty SWB with 24 vertical tubes. The standards were loaded at one of four vertical heights within the tube, 0 cm, 20.3 cm (8 inches), 40.6 cm (16 inches) and 61.0 cm (24 inches). Measurements within a range of plutonium loadings up to 75 g WG Pu (4.55 g Pu-240e) were taken with the neutron system to establish calibration parameters. For each Pu loading tested, 6 replicates measurements were acquired.
The SuperHENC neutron calibration is based on a uniform distribution of source material throughout the volume of the SWB (known as the “volume average” method) at each Pu mass source loading. The impact of deviations of the actual source position within a real waste box from the “volume average” calibration baseline is quantified in the TMU analysis (described later). It is not necessary to actually have a uniform source distribution at each calibration loading, because the chamber efficiency profile is known. Thus, the calibration measurements were performed with the calibration standard(s) in a single configuration. An efficiency correction factor was applied to convert the measured response (net doubles rate) at the calibration position to a “simulated” response for a volume average source distribution.

The calibration curve for Pu-240e was determined by a linear regression using the volume average corrected net doubles or net ones calibration data forced through the origin (zero grams of Pu-240e corresponds to zero net counts).

**Multiplicity Parameters**

Since the multiplicity analysis directly solves a system of three equations in three unknowns, a calibration curve per se is not required. Using data from the chamber efficiency measurements, the doubles gate fraction, and triples gate fraction were determined to be 0.628 and 0.457 respectively.

**Calibration Confirmation**

The neutron and gamma calibration was confirmed by assay of plutonium standards (different from the calibration standards) chosen as representative of the dynamic range of the system. The measurements were taken in five SWB surrogate boxes representing an empty box, light metals, plastics, dry combustibles and wet combustible wastes. These were measured in accordance with the routine operating instruction. Neutron assays take between 800 and 1800 seconds, with the measurement completing early if a specified level of precision 3% is met. The gamma assay comprises a 450 second real time gamma data acquisition period on each side of the box.

The calibration confirmation measurements are required to meet the WIPP waste acceptance criteria (WAC) specified in [1]. The results of the calibration confirmation measurements are presented in Table II and Table III. All measurements on non-interfering boxes met the specified acceptance criteria for %R and %RSD. The interfering box measurements all met the acceptance criteria for %R and %RSD stipulated under the DOE’s Performance Demonstration Plan (PDP) [2].

All of the above results were determined with standard “doubles mode” coincidence analysis with AAS matrix correction. The 30.1g Pu-240e confirmation measurements were also analyzed using the “solve for efficiency” multiplicity analysis method [4]. Comparison of the resulting Pu240e mass yielded the following conclusions regarding the relative merits of the two methods:

- The precision for the empty box in multiplicity mode is worse than in the standard doubles mode (11.0 %RSD compared to 0.4 %RSD).
- The accuracy for the empty box is about the same in both multiplicity mode and standard doubles mode (96.3 %R compared to 106.7 %R).

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1 The term “%R” is a measure of accuracy. It is the mean of the measured results as a percentage of the true (tag) mass.

2 The term “%RSD” is a measure of precision. It is the standard deviation in the measured results as a percentage of the true mass.
• The accuracy for the interfering boxes is better in multiplicity mode than in standard doubles mode\(^3\) (103.6 - 115.9 %R compared to 135.8 – 146.8 %R).

These conclusions follow the theoretical expectation for multiplicity mode i.e. that the precision is worse because the high order multiplicity results (e.g., triples) have poor precision and the accuracy is better with the interfering boxes because of the multi-parameter analysis.

It is recommended that expert review (including multiplicity mode analysis) be performed under the following conditions:

• AAS Correction Factor is greater than 2.2, AND
• Measured Pu-240e mass (AAS doubles mode) exceeds 15.2g Pu-240e (250g WG Pu).

Further confirmation of the linearity of the calibration for the empty was performed using the data for the chamber efficiency measurements with the Cf-252 source. This source has a high neutron emission rate (78782 n/s on 12/17/2004) and thus provides a good test of the linearity assumption in the doubles calibration line for high Pu loadings. The doubles emission rate is calculated by multiplying the spontaneous fission (SF) rate by the 2nd moment of SF for Pu-240 and dividing by two. For Cf-252, alpha is zero, therefore the singles neutron emission rate is simply the decayed total certified neutron output. The SF rate is then calculated by dividing this into the 1st moment of SF for Cf-252. This allows us to determine the doubles neutron emission rate for the Cf-252 source in the same manner used for the Pu standards.

Figure 3 shows the resulting plot of measured rate against emission rate for singles and doubles, where the measured rates are the calibration confirmation measurements and chamber efficiency measurements. This plot demonstrates the linearity of the Pu-240e to doubles calibration line up to and beyond the confirmed range (i.e. 30.1g Pu-240e).

Table II. Calibration Confirmation with Non-Interfering Matrices.

<table>
<thead>
<tr>
<th>Tag g Pu</th>
<th>Avg Meas g Pu</th>
<th>Tag alpha-Ci</th>
<th>Avg Meas alpha-Ci</th>
<th>WIPP-WAC %R (Pu)</th>
<th>WIPP-WAC %R (Ci)</th>
<th>WIPP-WAC %RSD (Pu)</th>
<th>WIPP-WAC %RSD (Ci)</th>
<th>PASS/FAIL</th>
<th>STATUS</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.2975</td>
<td>0.349</td>
<td>0.024</td>
<td>0.028</td>
<td>117.3%</td>
<td>118.9%</td>
<td>4.1%</td>
<td>4.2%</td>
<td>PASS</td>
<td></td>
</tr>
<tr>
<td>5.0430</td>
<td>6.315</td>
<td>0.406</td>
<td>0.512</td>
<td>125.2%</td>
<td>126.2%</td>
<td>2.9%</td>
<td>3.0%</td>
<td>PASS</td>
<td></td>
</tr>
<tr>
<td>80.0168</td>
<td>74.722</td>
<td>6.442</td>
<td>6.029</td>
<td>93.4%</td>
<td>93.6%</td>
<td>1.0%</td>
<td>1.0%</td>
<td>PASS</td>
<td></td>
</tr>
<tr>
<td>159.9600</td>
<td>166.125</td>
<td>12.943</td>
<td>13.330</td>
<td>103.9%</td>
<td>103.0%</td>
<td>1.1%</td>
<td>1.1%</td>
<td>PASS</td>
<td></td>
</tr>
<tr>
<td>249.9997</td>
<td>260.066</td>
<td>20.195</td>
<td>20.663</td>
<td>104.0%</td>
<td>102.3%</td>
<td>1.7%</td>
<td>1.6%</td>
<td>PASS</td>
<td></td>
</tr>
<tr>
<td>485.3542</td>
<td>516.429</td>
<td>39.420</td>
<td>41.134</td>
<td>106.4%</td>
<td>104.3%</td>
<td>1.6%</td>
<td>1.6%</td>
<td>PASS</td>
<td></td>
</tr>
</tbody>
</table>

\(^3\) It is believed that the high bias observed for interfering boxes will only manifest itself for boxes with low Pu-240 mass fraction isotopics (such as WG Pu) because the effect is due to multiplication which requires a high concentration of fissile material.
Table III. Calibration Confirmation with Interfering Matrices.

<table>
<thead>
<tr>
<th>Matrix</th>
<th>Tag Avg Meas Pu</th>
<th>Tag Avg Meas alpha-Ci</th>
<th>WIPP-WAC %R (Pu)</th>
<th>WIPP-WAC %R (Ci)</th>
<th>PASS/FAIL STATUS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Metals</td>
<td>5.0430</td>
<td>4.279</td>
<td>0.367</td>
<td>84.9%</td>
<td>90.5% PASS</td>
</tr>
<tr>
<td>Plastics</td>
<td>5.0430</td>
<td>4.024</td>
<td>0.334</td>
<td>79.8%</td>
<td>82.3% PASS</td>
</tr>
<tr>
<td>Dry Com</td>
<td>5.0430</td>
<td>4.958</td>
<td>0.389</td>
<td>98.3%</td>
<td>95.7% PASS</td>
</tr>
<tr>
<td>Wet Com</td>
<td>5.0430</td>
<td>4.494</td>
<td>0.361</td>
<td>89.1%</td>
<td>89.0% PASS</td>
</tr>
<tr>
<td>Metals</td>
<td>80.0168</td>
<td>84.310</td>
<td>6.442</td>
<td>105.4%</td>
<td>101.6% PASS</td>
</tr>
<tr>
<td>Plastics</td>
<td>80.0168</td>
<td>71.644</td>
<td>6.442</td>
<td>89.5%</td>
<td>93.2% PASS</td>
</tr>
<tr>
<td>Dry Com</td>
<td>80.0168</td>
<td>66.662</td>
<td>6.442</td>
<td>83.3%</td>
<td>82.0% PASS</td>
</tr>
<tr>
<td>Wet Com</td>
<td>80.0168</td>
<td>70.321</td>
<td>5.473</td>
<td>87.9%</td>
<td>85.0% PASS</td>
</tr>
<tr>
<td>Metals</td>
<td>159.9600</td>
<td>138.373</td>
<td>12.943</td>
<td>107.7%</td>
<td>104.2% PASS</td>
</tr>
<tr>
<td>Plastics</td>
<td>159.9600</td>
<td>157.947</td>
<td>12.943</td>
<td>98.7%</td>
<td>100.3% PASS</td>
</tr>
<tr>
<td>Dry Com</td>
<td>159.9600</td>
<td>205.668</td>
<td>12.943</td>
<td>128.6%</td>
<td>125.5% PASS</td>
</tr>
<tr>
<td>Wet Com</td>
<td>159.9600</td>
<td>172.232</td>
<td>13.481</td>
<td>107.7%</td>
<td>104.2% PASS</td>
</tr>
</tbody>
</table>

Fig. 3. Plot of SHENCA neutron calibration linearity confirmation at Hanford
ABSOLUTE GAMMA MODE CALIBRATION

Gamma calibration measurements were performed with line sources loaded into empty, dry combustibles and metals surrogate waste boxes. The absolute gamma efficiency was determined as a function of energy and matrix mass. A correction factor was applied to account for the “layering” effect of the matrix material in the metals surrogate which contained six layers of metal paint tins containing scrap metal.

Calibration confirmation analysis was performed for the absolute gamma calibration. This mode was tested for Pu-239 absolute gamma mass determination (from the 413.7 keV line) for interfering and non-interfering surrogate matrices over the range 0.48 to 455.3 g Pu-239. All CH-WAC criteria were met for %R and %RSD in the non-interfering SWB matrix up to 455.31 g Pu-239. For the interfering matrices, the PDP criteria were met for %R up to 455.31 g Pu-239. In addition the calibration was confirmed with the addition of the 10mm steel filter for the range 18.77 to 70.38 g Pu-239.

LOWER LIMIT OF DETECTION

The lower limit of detection (LLD) is defined as that level of radioactivity that, if present, yields a measured value greater than the critical level with a 95% probability, where the critical level is defined as that value which measurements of the background will exceed with 5% probability. In determining the LLD one must also account for interferences from different matrix conditions or radiation backgrounds that occur in the waste.

In order to perform TRU/LLW sorting, the system LLD must be less than 100 nCi of TRU alpha activity per gram of waste matrix. Furthermore, the Hanford buyer required that the system must be capable of a LLD of less than 60 nCi/g in a SWB with nominal lower net weight of 300 lb (136kg).

The routine assay measurement comprises a background measurement with an empty chamber followed by a measurement on the box itself. The net count rates are determined by subtracting the empty chamber background count rate from the gross count rate for the waste box. The empty chamber measurement provides an approximation for the true background count rate that one would have obtained if the waste box were free of radioactive material. With the SuperHENC neutron system, this background is a function of the matrix (materials present in the unknown waste box). For example, the presence of high Z materials (e.g. Fe) may increase the background relative to the empty chamber due to cosmic spallation, whereas the presence of neutron absorbers such as hydrogen will have the opposite effect.

The background count rates are not a simple function of matrix mass. The background is reduced by the presence of organic material (e.g. for the plastics and combustibles) and increased by the presence of metals (e.g. with the metals SWB). Presumably the former effect is caused by the neutron absorption in hydrogen and the latter effect is due to increased cosmic spallation in the high Z metals. As Hanford do not intend to segregate metals from the organic materials, there will be no way to reliably correct the background of an unknown waste item using this data. Therefore it is assumed that the background is “flat” i.e. that the background measured with an empty chamber will be the same as the background with the waste item. This is a reasonable assumption, because the waste is likely to contain a mixture of metals and organics in which the two effects described above will cancel out for most waste items.

Various methods are used by the SuperHENC to reduce the background and the variance therein:

- A specially designed veto circuit is used to filter out local coincidences which are more likely to be the result of cosmic spallation than due to spontaneous fission from TRU nuclides within the SWB. For a period of 128 microseconds after a neutron event is counted in a particular bank of detectors, no further neutrons are counted in that bank and its two neighboring banks. At high
count rates, the veto must be turned off because it reduces the chamber efficiency. For routine assays, the software performs a preliminary (10 second) measurement to determine if the veto should be turned on or off.

- The truncated multiplicity (ones) method [4] is applied at low count rates to diminish the effect of high order multiplicity events that are again usually the result of cosmic spallation.

It was found that the truncated multiplicity method reduces the background by 25%, and the use of the veto further reduced the background by 1%. At Hanford’s 1,000 ft elevation, the cosmic background is significantly lower than at high elevation sites such as Rocky Flats or Los Alamos. As a result, the veto circuitry plays a less important roles in reducing the LLD compared to high elevation sites.

Both background reduction methods involve loss of some signal as well as noise, so the threshold for their use was set at low count rates. For the veto, the threshold was set to at a net singles count rate of 15 c/s and for the truncated multiplicity the threshold was set at a net doubles rate of 32 c/s.

The two factors that have the largest impact on the Hanford SuperHENC neutron LLD are:

- the statistical variance in the true neutron background for the unknown waste box - this component will be referred to as the “A” term,
- the systematic variance between the estimated background (derived from the empty chamber measurement) and true background for the unknown waste matrix - this component will be referred to as the “F” term.

The magnitude of the systematic variance in background due to matrix, $LLD_F$, was estimated by calculating the (apparent) average Pu-240e mass induced by the presence of the blank (defined as a simulated waste matrix that contains no added activity) surrogate matrix in the chamber. $LLD_F$ (units of g Pu-240e) was found to be dependent on the mass, $m$, of the waste as follows:

$$LLD_F = 3.5 \times 10^{-3} m$$  \hspace{1cm} (Eq. 3)

To quantify the A-term of the LLD, eight replicate measurements of a full (0% void), blank matrix (no Pu loaded) were collected for five SWB surrogate matrices. The replicates comprised a no-source assay measurement with a surrogate matrix box paired with an empty chamber background measurement. For each matrix, $LLD_A$ was determined by applying group statistics to a sample of repeat assays carried out on blank waste matrices. From this data, the following relationship between net mass and $LLD_A$ (units of g Pu-240e), has been determined:

$$LLD_A = 2.5 \times 10^{-3} + 4.1 \times 10^{-6} m$$  \hspace{1cm} (Eq. 4)

The minimum detectable concentration (MDC) in terms of nCi of TRU alpha activity per gram of waste matrix is defined as:

$$MDC = 10^6 \frac{Act_{spec}}{m} \sqrt{LLD_A^2 + LLD_F^2}$$  \hspace{1cm} (Eq. 5)

where $Act_{spec}$ is the specific TRU alpha activity of the isotope or mixture of interest (Ci/g).
The relationship between LLD (in terms of g WG Pu) and MDC (nCi/g for WG Pu) is plotted in Figure 4. We see that LLD reaches its maximum value for the highest matrix mass, whereas MDC reaches its maximum value for the lowest matrix mass. Note that for 12% and 18% Pu-240/Pu isotopics, both the LLD and the MDC is lower than for the 6% Pu-240/Pu (WG) isotopics.

Absolute Gamma LLDs have been determined for matrix masses up to 544 kg for the major photons emitted from U-235, U-233, Cs-137 and U-238 (via Pa-234m progeny). The absolute gamma mode LLD for U-235 has been demonstrated to be less than 0.5 g for SWBs containing up to 544 kg matrix. In addition the LLD has been determined for measurement of Pu-239 via the 413.7 keV photon although it is envisaged that plutonium assay would be in most cases performed with the neutron system. It has been demonstrated that TRU/LLW sorting may be performed for WG Pu in absolute gamma mode.

![Graph showing LLD and MDC plotted against net matrix mass](image-url)
TOTAL MEASUREMENT UNCERTAINTY (TMU)

All significant sources of uncertainty have been quantified including random and systematic effect associated with the neutron measurement and isotopic effects associated with the integration of neutron data with the PC-FRAM gamma analysis. The following independent sources of measurement uncertainty are combined in quadrature to determine the TMU:

- **Random effects** - introduced by counting statistics. The relative statistical uncertainty component (or “precision”) in the Pu-240e mass is calculated in the software.

- **Matrix effects** – due to impact of heterogeneous waste forms as a deviation from calibration baseline. Matrix uncertainty was estimated using data acquired with surrogate matrices constructed to simulate 0%, 15%, or 30% by volume void space. The uncertainty is dependent on CF and is best expressed as follows:
  \[
  U_{\text{Mat}} = 0.1015(CF - 1) + 0.03 \\
  = 0.03 
  \]
  \[(\text{Eq. 6})\]
  \[CF > 1\]
  \[CF \leq 1\]

- **Source position effects** – due to the variation of source position within the box as a deviation from the calibration baseline (uniform distribution). This effect is dependent on CF and is best characterized by the following expression:
  \[
  U_{\text{Pos}} = 0.08(CF - 1) + 0.017 \\
  = 0.017
  \]
  \[(\text{Eq. 7})\]
  \[CF > 1\]
  \[CF \leq 1\]

- **Calibration effects** – due to differences in the physical properties of the sources used for calibration, uncertainties associated with the source activity and uncertainties that arise in the curve fitting and position average correction processes. This term is estimated to be +/- 1%.

- **Background effects** – due to variation between the estimated and actual background. This is estimated to be +/- 15% for less than or equal to 3 g WG Pu. For assays above this level, the uncertainty in the background contribution is small compared to the gross signal.

- **Multiplication effects** – generate a high bias in the reported mass for large concentrated lumps of plutonium. This is estimated at 1.5 % for assay results greater than 100g Pu and zero otherwise.

The TMU in total Pu is evaluated in Table IV. This includes an uncertainty contribution to account for isotopic uncertainties for the conversion from Pu-240e to the parameter of interest.
Table IV. TMU at Various Pu Loadings and Various AAS CF Values.

<table>
<thead>
<tr>
<th>WG Pu Mass (g)</th>
<th>Pu240e Mass (g)</th>
<th>Total Pu TMU % At Various AAS CF</th>
<th>1.0</th>
<th>1.9</th>
<th>2.8</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0.006</td>
<td></td>
<td>23.3%</td>
<td>&lt;LLD</td>
<td>&lt;LLD</td>
</tr>
<tr>
<td>0.2</td>
<td>0.012</td>
<td></td>
<td>18.9%</td>
<td>23.9%</td>
<td>32.5%</td>
</tr>
<tr>
<td>0.3</td>
<td>0.018</td>
<td></td>
<td>17.2%</td>
<td>22.6%</td>
<td>31.6%</td>
</tr>
<tr>
<td>0.5</td>
<td>0.030</td>
<td></td>
<td>16.5%</td>
<td>22.1%</td>
<td>31.2%</td>
</tr>
<tr>
<td>1</td>
<td>0.061</td>
<td></td>
<td>34.0%</td>
<td>21.7%</td>
<td>30.9%</td>
</tr>
<tr>
<td>2</td>
<td>0.12</td>
<td></td>
<td>29.5%</td>
<td>36.9%</td>
<td>30.8%</td>
</tr>
<tr>
<td>3</td>
<td>0.18</td>
<td></td>
<td>20.9%</td>
<td>34.2%</td>
<td>40.7%</td>
</tr>
<tr>
<td>5</td>
<td>0.30</td>
<td></td>
<td>16.5%</td>
<td>29.8%</td>
<td>37.1%</td>
</tr>
<tr>
<td>10</td>
<td>0.61</td>
<td></td>
<td>15.9%</td>
<td>29.0%</td>
<td>37.0%</td>
</tr>
<tr>
<td>80</td>
<td>4.86</td>
<td></td>
<td>15.4%</td>
<td>28.2%</td>
<td>36.9%</td>
</tr>
<tr>
<td>250</td>
<td>15.19</td>
<td></td>
<td>9.2%</td>
<td>23.4%</td>
<td>32.5%</td>
</tr>
<tr>
<td>500</td>
<td>30.38</td>
<td></td>
<td>8.6%</td>
<td>23.0%</td>
<td>32.4%</td>
</tr>
</tbody>
</table>

PDP RESULTS

The results of the “blind” PDP tests for cycle B5A performed on June 2005 with SHENCA are indicated in Table V. The system passed these tests for all matrix/source combinations.

Table V. PDP Results for SHENC at Hanford for Cycle B5A.

<table>
<thead>
<tr>
<th>SWB Matrix</th>
<th>%RSD</th>
<th>%R</th>
<th>STATUS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Non-Interfering</td>
<td>2.55</td>
<td>91.34</td>
<td>PASS</td>
</tr>
<tr>
<td>Combustibles</td>
<td>0.81</td>
<td>79.55</td>
<td>PASS</td>
</tr>
<tr>
<td>Metals</td>
<td>6.38</td>
<td>120.21</td>
<td>PASS</td>
</tr>
</tbody>
</table>
SUMMARY

A new SuperHENC assay system has been commissioned at Hanford for assay of heterogeneous debris waste in SWBs. New software has been developed for integration of neutron and gamma data. The system has been calibrated in neutron assay mode (with PC-FRAM / AK isotopic analysis) and in absolute gamma mode. The calibrations have been confirmed with assays performed using routine operating procedures with special nuclear materials. The lower limits of detection and total measurement uncertainty have been determined. Table VI summarizes the qualified ranges. It has been demonstrated that the new system is meets all applicable regulatory performance objectives and provides for TRU/LLW sorting of un-segregated debris waste with diverse isotopic mixtures.

Table VI. Summary of Hanford SuperHENC Performance.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Neutron Assay Mode with PC-FRAM</th>
<th>Absolute Gamma Mode</th>
</tr>
</thead>
<tbody>
<tr>
<td>Container / Matrix:</td>
<td>SWBs / Heterogeneous Debris Waste</td>
<td>SWBs / Heterogeneous Debris Waste</td>
</tr>
<tr>
<td>Qualified Pu Range:</td>
<td>Pu-240e: LLD – 30.1g WG Pu: LLD – 502</td>
<td>Pu-239: LLD – 445.31 g</td>
</tr>
<tr>
<td>Qualified Matrix Range:</td>
<td>AAS CF: 0.963 – 3.107 Waste mass: 0 – 1925.7 kg</td>
<td>Waste mass: 0 – 550 kg</td>
</tr>
<tr>
<td>Expert review range:</td>
<td>CF &gt;2.2 and Pu-240e: 15.2 - 30.1 g</td>
<td>N/A</td>
</tr>
<tr>
<td>LLD (g):</td>
<td>Pu-240e: 0.0025 – 0.0683 g</td>
<td>Pu-239: 0.12 – 0.40 g</td>
</tr>
<tr>
<td></td>
<td></td>
<td>U-235: 0.07 – 0.28 g</td>
</tr>
<tr>
<td>MDC (nCi/g)</td>
<td>51.5 (WG Pu, 280 kg waste mass)</td>
<td>90.15 (WG Pu, 280 kg waste mass)</td>
</tr>
<tr>
<td>TMU</td>
<td>16.5% - 37.1% (for 5g WG Pu)</td>
<td>39.9% - 62.3% (for 5g WG Pu)</td>
</tr>
</tbody>
</table>
REFERENCES