Performance Assessment of the Add-A-Source Matrix Correction Method Using the Super High Efficiency Neutron Coincidence Counting System (SuperHENC) - 10527

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ABSTRACT

In January 2001, the Super High Efficiency Neutron Counter (SuperHENC) was the first boxed waste assay system in the DOE complex to be certified for disposal to the Waste Isolation Pilot Plant (WIPP). Over the past decade, three systems have been built, performing high sensitivity characterization at various sites. The SuperHENC technique has now become a routine method for characterization of large containers.

Key to the success of the SuperHENC is the robust Add-A-Source (AAS) matrix correction method. A small Cf-252 source is placed into the assay chamber to acquire neutron coincidence data with and without the waste container loaded. This ratio allows a correction factor to be applied to the measured Pu240 effective mass result for matrix interferences.

Using the large amount of data available from thousands of containers measured to date, a study has been undertaken to analyze the performance of the AAS method in order to (i) assist in predicting the likely range of AAS correction factors for commonly encountered waste streams (ii) assess the magnitude of measurement uncertainty as a function of matrix content (iii) ensure that appropriate matrix surrogates can be constructed that are appropriate simulations of anticipated waste streams, and (iv) determine the bounding limits on the AAS technique when applied to boxed waste.

The results of the study demonstrate the versatility and flexibility of the SuperHENC platform in characterizing a diverse range of matrix materials. Waste forms such as combustibles, plastics, metals, compacted pucks, and sludge have been successfully measured to date.

INTRODUCTION

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

The SuperHENC has, to date, been deployed at four locations in the United States Department of Energy (DOE) complex. Table I summarizes the deployment and certification history against WIPP performance requirements [1, 2].

Table I. SuperHENC Deployment Sites

Site	Purpose	Waste Streams	WIPP Certification
Rocky Flats Environmental Technology Site (RFETS)	Waste Characterization	Heterogeneous debris waste in SWBs	January 2001
Hanford WRAP	Waste Characterization	Waste Receiving and Processing (WRAP) debris waste in SWBs	June 2005
Hanford PFP	Safeguards	Plutonium Finishing Plant (PFP) debris waste in SWBs	N/A
Idaho National Laboratory (INL).	Waste Characterization	100-gallon drums containing compacted 55-gallon waste drums (pucks), SWBs containing drums of sludge and SWBs containing debris waste	February 2007.

SYSTEM DESCRIPTION

The SuperHENC is a passive neutron counter combined with high resolution gamma spectrometer. The neutron counter consists of arrays of He-3 detectors embedded in all six sides of the neutron counting chamber thus providing a high efficiency 4π neutron detector. The gamma spectrometer consists of a single High Purity Germanium (HPGe) detector and a turntable to allow viewing different sides of the SWB. The turntable also serves as a scale for weighing the SWB during the gamma measurement.

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.

SuperHENC measures the Pu-240e content using passive neutron time-correlation counting and calculates the total plutonium content using either Acceptable Knowledge (AK) or direct gamma measurement for the plutonium isotopic mass fractions (Pu-238, Pu-240 and Pu-242 are the only significant spontaneous fission source in Pu waste streams) and other radionuclides present.

The neutron system runs under a derivative of the Los Alamos National Laboratory (LANL) General Purpose Neutron Coincidence Counting (INCC) software [5]. The neutron coincidence circuitry uses the Advanced Multiplicity Shift Register (AMSR 50) which is fully supported by the INCC program. The SWB is first loaded on the gamma turntable using a fork lift fitted with a special handling attachment where it is weighed and measured with the gamma spectrometer. Then it is transferred to the neutron chamber where neutron assay is performed. Using the gamma spectrum, relative Pu isotopic and other nuclide fractions are determined which then are folded using PC-FRAM [6] with the neutron data to produce a final radioassay report.

Matrix Correction

The neutron counter uses the Add-A-Source (AAS) method [7] for performing matrix correction and normalization. The AAS is a Cf-252 source attached to a Teleflex^{TM} cable that travels a serpentine path under the neutron assay chamber, stopping at six pre-selected positions. When not in use, the source is retracted from the chamber and stored in a polyethylene pig.

The INCC software calculates the measured response to the AAS, compares this to a reference count and calculates the matrix correction factor. The normalization is a simple and quick check on the empty neutron chamber counting efficiency compared to a reference initial source measurement.

Calibration

The calibration comprises several steps. These elements include 1) mapping chamber efficiency with a neutron source, 2) setting up the AAS, 3) constructing a Monte Carlo model [8,9] 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.

Monte-Carlo Model

A Monte Carlo N-Particle (MCNP) model [8, 9] was used to determine the relationship between AAS response and matrix correction factor. Various waste compositions and densities were modeled for the system. The matrix loadings that were modeled included polyethylene with density variation of 0, 0.015, 0.030, 0.050 and 0.100 g/cm^3 and iron at densities 0, 0.130, 0.220, 0.450 and 0.670 g/cm^3. The AAS matrix correction factor (CF) is calculated from the following expression:

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

where *a*, *b*, *c*, and *d* are calibration coefficients determined from MCNP. For SWBs these coefficients have been determined to be *a*= -5.1366E-02, *b*=2.7148E-01, *c*=2.566E+00 and *d*= -5.201E-01.

$$\Delta = r - 1 \qquad \text{(Eq. 2)}$$

where *r* is the ratio of the (decay corrected) AAS reference doubles rate (i.e., with empty box) to the measured AAS 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 AAS 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 at each site.

EXPERIENCE AT RFETS

At RFETS, the SuperHENC needed to comply with WIPP and RFETS Nuclear Material Control and Accountability (MC&A) performance requirements for accuracy and precision. The criteria that had been originally developed for 55-gallon (208 liter) drums were applied to large boxes as a conservative requirement. The system was operated at RFETS for four years and was a key component in the shipment of over 4,000 SWBs to WIPP.

Nuclear Materials Control and Accountability Requirements

The RFETS MC&A requirements for precision and accuracy at RFETS are summarized in Table II. Validation measurements were collected on a non-interfering matrix (empty) standard for WIPP method performance demonstration, and on a variety of surrogate matrices that were representative of the waste stream at RFETS. Six replicates on three Pu loadings (1, 10, and 320 g) were collected [10]. Sources were located in the approximate volume average position. This Pu range characterized the entire expected range, from lower detection limit to the SWB criticality-loading limit.

Table II. MC&A Quality Assurance Objectives for Radioassay at RFETS

Range of Pu and U (g)	Precision (%) [a]	Accuracy (%) [b]
≤1.0	50	50 to 150
>1.0 to ≤10	25	75 to 125
>10.0	10	85 to 115

[a] Ratio of standard deviation in measured values of the known value, expressed as a percent
[b] Limits on the two-sided 95 percent confidence bound for the ratio of the mean of the measured values to the known value, expressed as a percent.

The surrogate matrices comprised SWBs loaded with a modular matrix cube design representative of RFETS materials such as mixed metals, dry combustibles and plastics. RFETS segregated its waste streams into well defined “item description codes” (IDCs) such as mixed metals, dry combustibles, plastics and mixed matrix IDCs defined as:

- Inorganic matrices with less than 10% by weight of organics (IDC 3010).
- Inorganic matrices with greater than 10% by weight of organics (IDC 3011).

The benefit of looser segregation criteria was reduced human exposure, cost and improved schedule. To meet MC&A qualification requirements, mock up standards of IDC 3010 and 3011 were constructed by combining cubes of metal and plastics to achieve 10% and 30% by weight organic content respectively.

Table III gives a summary of the SuperHENC measurements taken at RFETS on independent Pu standards to validate the calibration [11]. Data were collected with the standard operating procedure and assay parameters used in routine operations. All of the measurements included in Table III passed the applicable WIPP and MC&A data quality objectives for SWBs based on %R[1] and %RSD[2].

[1] %R is a measure of accuracy, i.e. the mean of the measured results as a percentage of the true (tag) mass.
[2] %RSD is a measure of precision, i.e. the standard deviation in the measured results as a percentage of true mass.

Table III. Validation Measurement Summary for SWBs at RFETS

Matrix (IDC)	0.9g WG Pu		9.0g WG Pu		320g WG Pu	
	%R	%RSD	%R	%RSD	%R	%RSD
Metals (480)	140	6.4	109	4.1	102	0.9
Mixed (3010)	110	3.5	94	1.0	99	0.8
Mixed (3011)	92	4.0	95	0.8	102	1.1
Plastics (337)	N/A	N/A	96	2.1	102	2.2
Zero (000)	126	13.4	106	1.5	103	0.6

Lower Limit of Detection

The SuperHENC's lower limit of detection (LLD) was determined from background measurements at RFETS. Instruments performing TRU/LLW discrimination measurements must have an LLD or minimum detectable concentration (MDC) less than 100 nCi/g. MDC is defined as that radioactivity concentration which, if present, yields a measured value greater than the critical level with 95% probability, where the critical level is defined as that value which measurements of the background will exceed with 5% probability. For the SuperHENC, MDC has been determined, for a homogenous spatial distribution of the source material, by statistical analysis of replicate assays of blank waste matrices (i.e. containing no added activity).

The MDCs for RFETS are summarized in Table IV for various surrogate matrices. These were calculated using RFETS weapons grade Pu isotopics (including approximately 30 year Am-241 in-growth) and the net weight of the matrix. The minimum detectable activity (MDA) in terms of WGPu mass is also shown in Table IV.

Table IV. Detection Limit Summary for the SuperHENC at RFETS

Matrix (IDC)	Matrix weight (kg)	MDA (g WGPu)	MDC	
			(nCi/g)	(Bq/g)
Empty (000)	0	0.113	N/A	N/A
Metals (480)	599	0.193	25.8	955
Dry Combustibles (330)	300	0.183	48.8	1806
Mixed (3010)	535	0.219	29.2	1080
Mixed (3011)	449	0.159	28.3	1047

Performance Demonstration Plan (PDP) Results

The first two cycles of the SWB PDP were conducted in May 2001 and April 2002. A combustible and a stainless steel matrix were tested. Six replicates of each sample were taken using the SuperHENC system and integrated in accordance with the standard operating procedure. The final scoring results reported by

the PDP for these two cycles are presented in Table V. All measurements passed the PDP SWB cycle acceptance criteria.

Table V. SuperHENC PDP Cycles Summary for SWBs

PDP Box Cycle #	Matrix (IDC)	Pu loading (g WG Pu)	PDP Score	
			%R	%RSD
B1A	Combustibles (330)	7	98	2.8
B1A	Metals (480)	10	106	2.0
B2A	Combustibles (330)	8	91	5.2
B2A	Metals (480)	7	100	2.7

Summary of RFETS Results

The SuperHENC was successfully calibrated and validated at RFETS [10, 11]. All MC&A and WIPP criteria for assay of 55-gallon drums were met for the SWBs. The qualification of the mixed-matrix waste streams crucial to the successful D&D of RFETS represented a new milestone in waste management. PDP cycle data also met acceptance criteria. A detailed MDC assessment was performed and demonstrated its suitability for TRU/LLW sorting at the 100nCi/g level (3700 Bq/g). The combined SuperHENC system achieved WIPP certification and was in full-time operation, measuring over 4,000 SWBs at RFETS from 2001 – 2005.

Certification of SuperHENC against WIPP and MC&A requirements was a significant technical achievement given the relevant data quality objectives that were originally developed for 55 gallon (208 liter) drums.

EXPERIENCE AT HANFORD

Several new challenges were faced with the SuperHENC systems that were built for deployment at Hanford:

- The original RFETS system was calibrated for segregated waste streams such that metals, plastics, wet combustibles and dry combustibles were separated by "Item Description Code" prior to assay.
- The RFETS mission of handling only Weapons Grade Pu enabled the original SuperHENC to benefit from the use of known Pu isotopics. Operations at Hanford (and most other DOE sites) generate non-segregated waste streams, with a wide diversity of Pu isotopics.
- Consequently, the Hanford SuperHENCs were required to deal with the challenges presented by un-segregated waste matrices and also perform a real-time determination of isotopic grade for each box.
- 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 neutron system's software and calibration methodology were modified to encompass these new requirements. Performance was rigorously tested and validated against WIPP quality requirements. These modifications together with the mobile platform make the SuperHENC far more robust to handle diverse waste streams and allow for rapid redeployment around the DOE complex.

A new software package (NGI) was developed that integrates the neutron and gamma data to provide a final analysis report.

One system (SHENCA) was deployed at the Waste Receiving and Processing (WRAP) facility to measure TRU heterogeneous debris waste for sentencing to the Waste Isolation Pilot Plant (WIPP). A second identical system (SHENCB) was deployed at the Plutonium Finishing Plant (PFP) at Hanford in support of the site's Materials Control and Accountability (MC&A) program. These systems are depicted in Figure 1.

The calibration program of the WRAP SuperHENC consisted of extensive calibration measurements, calibration confirmation measurements with various matrices and Pu gram loadings, and Total Measurement Uncertainty (TMU) and Lower Limit of Detections (LLD) measurements. The calibration program which started in November 2004 was completed in March 2005. The calibration covered a range of LLD to 650 grams of WGPu. The LLD and consequently the MDC are functions of the net matrix weight. The WRAP SuperHENC went through initial WIPP Certification Audit and WIPP Performance Demonstration Program (PDP) and started assaying waste in June 2005.

(a) System Ready for Transportation

(b) Loading an SWB onto Gamma Turntable

(c) Neutron Assay Chamber

(d) SWB Surrogate Matrix

Fig. 1. Hanford SuperHENC system

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 [12, 13]. 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 3% precision was met.

The calibration confirmation measurements were required to meet the WIPP waste acceptance criteria (WAC). The results of the calibration confirmation measurements are presented in Table VI and Table VII. The measurements all met the acceptance criteria for %R and %RSD stipulated under the DOE regulations.

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. 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).
- The accuracy for the interfering boxes is better in multiplicity mode than in standard doubles mode (103.6 - 115.9 %R compared to 135.8 – 146.8 %R). 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.

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.

Table VI. Calibration Confirmation with Non-Interfering Matrices.

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

Table VII. Calibration Confirmation with Interfering Matrices.

Matrix	Tag g Pu	Avg Meas g Pu	Tag alpha-Ci	Avg Meas alpha-Ci	WIPP-WAC %R (Pu)	WIPP-WAC %R (Ci)	PASS/FAIL STATUS
Metals	5.0430	4.279	0.406	0.367	84.9%	90.5%	PASS
Plastics	5.0430	4.024	0.406	0.334	79.8%	82.3%	PASS
Dry Com	5.0430	4.958	0.406	0.389	98.3%	95.7%	PASS
Wet Com	5.0430	4.494	0.406	0.361	89.1%	89.0%	PASS
Metals	80.0168	84.310	6.442	6.547	105.4%	101.6%	PASS
Plastics	80.0168	71.644	6.442	6.007	89.5%	93.2%	PASS
Dry Com	80.0168	66.662	6.442	5.285	83.3%	82.0%	PASS
Wet Com	80.0168	70.321	6.442	5.473	87.9%	85.0%	PASS
Metals	159.9600	138.373	12.943	10.979	86.5%	84.8%	PASS
Plastics	159.9600	172.232	12.943	13.481	107.7%	104.2%	PASS
Dry Com	159.9600	205.668	12.943	16.368	128.6%	126.5%	PASS
Wet Com	159.9600	157.947	12.943	12.986	98.7%	100.3%	PASS
Metals	249.9997	265.128	20.195	20.684	106.1%	102.4%	PASS
Plastics	249.9997	276.921	20.195	22.766	110.8%	112.7%	PASS
Dry Com	249.9997	279.862	20.195	21.846	111.9%	108.2%	PASS
Wet Com	249.9997	315.549	20.195	25.955	126.2%	128.5%	PASS
Metals	485.3542	450.727	39.420	37.222	92.9%	94.4%	PASS
Plastics	485.3542	579.358	39.420	46.116	119.4%	117.0%	PASS
Dry Com	485.3542	612.244	39.420	49.604	126.1%	125.8%	PASS
Wet Com	**485.3542**	**665.662**	**39.420**	**54.898**	**137.1%**	**139.3%**	**CONDITIONAL PASS**

PDP Results

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

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

SWB Matrix	%RSD	%R	STATUS
Non-Interfering	2.55	91.34	PASS
Combustibles	0.81	79.55	PASS
Metals	6.38	120.21	PASS

EXPERIENCE AT IDAHO

Following the closure of the RFETS site, Pajarito Scientific Corporation (PSC) refurbished the original SuperHENC for deployment at Idaho National Laboratory (INL) for assay of 100-gallon drums containing compacted 55-gallon waste drums (pucks), SWBs containing drums of sludge and SWBs containing debris waste. Original certification for the SuperHENC system was issued in February 2007. Soon thereafter, the Environmental Protection Agency (EPA) added Tier 1 approval to the SuperHENC's impressive list of credentials.

Figure 2 shows the range of waste streams that have been encountered at INL using a plot of Gross Weight of the container against Matrix Correction Factor (CF). The compacted pucks and SWBs with debris waste dominate the low end of the CF region. Sludge demonstrates a wide range of CF due to the variable hydrogen concentration (water content). Also shown on this plot are some of the surrogate

matrices that have been built to simulate waste (including the surrogates – dry and wet combustibles, metals and plastics – used at Hanford). These are labeled 'heterogeneous surrogates' in Figure 2.

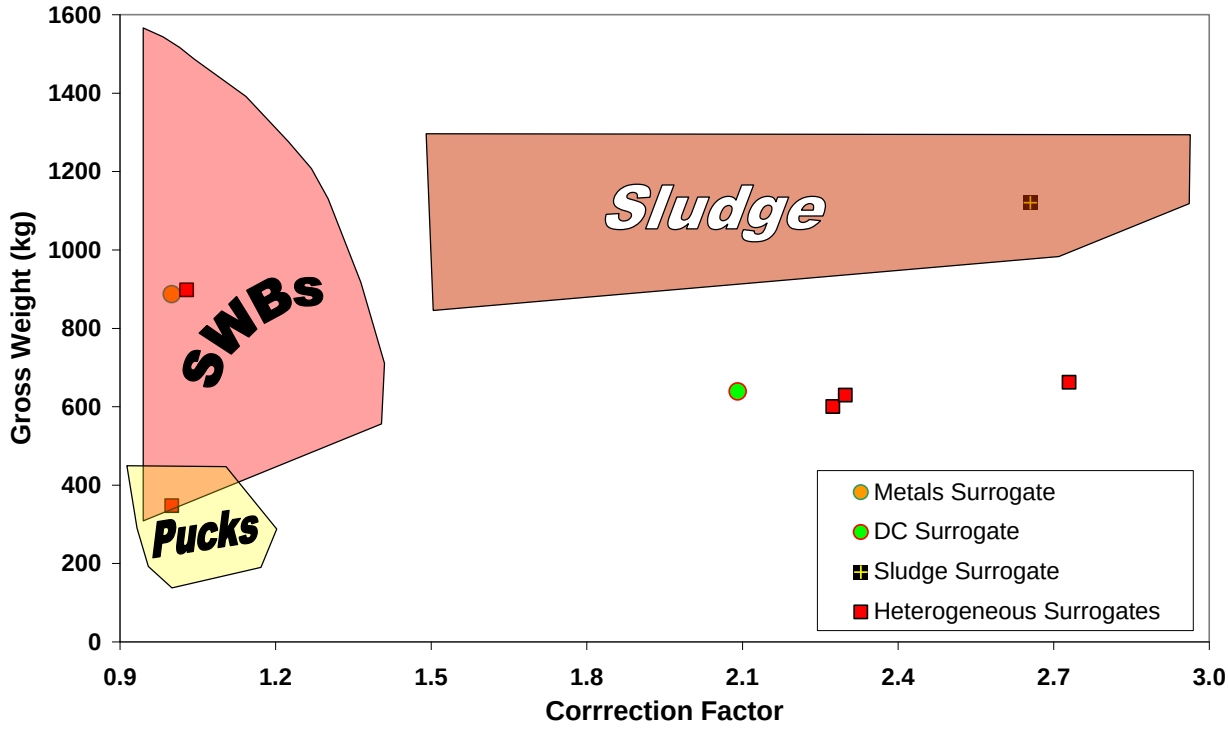

Figure 2. Range of Waste Streams at INL (Gross Weight compared to Matrix Correction Factor)

Interfering Matrix (IM) measurements are used "to assess the long-term stability of the NDA instrument's matrix correction" as required in the WAC. IM measurements for each surrogate matrix configuration are measured with appropriate source loadings to verify the operational range of the instrument during each six-month period. A surrogate interfering matrix container has been selected to reflect each waste configuration rather than using multiple configurations to cover a range of production waste matrices.

An SWB containing a light metals matrix was selected to reflect the 100-gallon puck drums, with the expectation that the major mass component of the puck drums will be the metal from the 55-gallon drums. A surrogate sludge matrix consisting of four sludge matrix drums placed in the SWB was selected to reflect the configuration of production sludge drums in an SWB. An SWB containing a dry combustible matrix was selected to represent the SWB debris waste. Three plutonium source-loading configurations were used for each interfering matrix spanning the range of calibration with approximate contents of 8, 106 and 203 grams of Total WG Pu. Results of the most recent IM runs are provided in Table IX.

The metals IM surrogate indicates good agreement with the production drums. On the average, the CF for the puck drums tended to center around CF=1.00. This agreement was expected since there was little uncertainty in neutron interferences associated with the metals configuration.

For the sludge surrogate, the assumption was that the sludge material would be wetter (more hydrogen) than was actually seen in the current production runs. However, as seen in Figure 2, some sludge SWBs have had higher CFs indicating the surrogate has a good overall indication of the possible neutron interferences.

Table IX. IM Results for SuperHENC

Container	Matrix	Avg CF	Max CF	Min CF
100G Puck Drums	Production	1.0643	1.1540	0.9670
100G Puck Drums	Surrogate	0.9984	1.0000	0.9430
Sludge SWBs	Production	2.5099	2.9240	2.1050
Sludge SWBs	Surrogate	2.9210	2.9830	2.8590
Debris SWBs	Production	1.0517	1.5650	0.9580
Debris SWBs	Surrogate	1.8966	1.9230	1.8610

The debris SWBs were a single-Site production run shipped from outside INL. Radiography indicated these SWBs contained high levels of metals in association with minor contributions of neutron interfering material such as plastics. While the surrogate is indicative of the overall possibilities in debris of non-segregated waste, it was not representative of this particular population containing mostly metals.

PDP Results

The results of the Idaho "blind" PDP tests for cycle B8A performed in August 2008 with SuperHENC are indicated in Table X. The system passed these tests for all matrix/source combinations.

Table X. PDP Results for SHENC at Idaho for Cycle B8A.

SWB Matrix	%RSD	%R	STATUS
Combustibles	1.72	104.6	PASS
Metals	1.18	106.4	PASS

SUPERHENC PERFORMANCE ASSESSMENT

The overall system can be best understood by examining the Total Measurement Uncertainty (TMU) of the system. All significant sources of uncertainty must be quantified including the random and systematic effect associated with the neutron measurement and the isotopic effects. Using data acquired to date from all DOE operating sites, the following independent sources of measurement uncertainty have been estimated for the SuperHENC system. The following terms are combined in quadrature to determine the final TMU in each measurement:

- **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 at Hanford [13] 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:

$$\begin{aligned} U_{Mat} &= 0.1015(CF-1)+0.03 \qquad CF>1 \\ &= 0.03 \qquad\qquad\qquad\qquad CF\le 1 \end{aligned} \qquad \text{(Eq. 3)}$$

- **Source position effects** – due to the variation of source position within the box as a deviation from the calibration baseline (uniform distribution). The deviation in detection efficiency for heterogeneously distributed Pu compared to the uniform distribution baseline is small because the SuperHENC has been designed to minimize variation in efficiency across the volume of the chamber for a diverse range of mixed matrix materials [9]. Source position uncertainty was estimated at Hanford [13] by examining the variation in detection efficiency for sources randomly positioned across the volume of the SWB. The uncertainty term is best characterized by the following expression:

$$\begin{aligned} U_{Pos} &= 0.08(CF-1)+0.017 \qquad CF>1 \\ &= 0.017 \qquad\qquad\qquad\quad CF\le 1 \end{aligned} \qquad \text{(Eq. 4)}$$

- **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. Note that, although we expect some degree of background uncertainty dependence on matrix, it is more appropriate to define a fixed background term than attempt to quantify the background uncertainty term as a function of CF. This is because the dominant source of matrix dependent background arises from cosmic spallation in metals which are transparent to the matrix correction.

- **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 performance of the AAS correction process is reflected here in terms of its contribution to the overall total measurement uncertainty. Source position effects and matrix effects are the dominant terms with source effect dominating at low CF (e.g. for metals) and the matrix term dominating for the high CF matrices such as sludge. The TMU in the Pu40 effective mass (Pu240e) at various Pu loadings for a typical SuperHENC system is evaluated in Table XI based on the study performed at Hanford [13].

Table XI. TMU at various Pu loadings and various AAS CF values.

WG Pu Mass (g)	Pu240e Mass (g)	Pu240e TMU% At Various AAS CF		
		1.0	1.9	2.8
0.1	0.006	23.3%	<LLD	<LLD
0.2	0.012	18.9%	23.9%	32.5%
0.3	0.018	17.2%	22.6%	31.6%
0.5	0.030	16.5%	22.1%	31.2%
1	0.061	16.1%	21.7%	30.9%
2	0.12	15.7%	21.5%	30.8%
3	0.18	4.7%	21.5%	30.8%
5	0.30	4.7%	21.5%	30.8%
10	0.61	4.2%	21.4%	30.7%
80	4.86	3.9%	21.3%	30.6%
250	15.19	4.0%	21.3%	30.6%
500	30.38	3.9%	21.3%	30.6%

Sensitivity

Another important performance consideration to consider the impact of the matrix correction process upon is the lower limit of detection. Background count rates are not a simple function of matrix mass. The background is reduced by the presence or organic material (e.g. for the plastics and combustibles) and increased by the presence of metals (e.g. with the metals SWB). The former effect is caused by the neutron absorption in hydrogen and the latter effect is due to increased cosmic spallation in the high atomic mass metals.

As many sites 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 approximately cancel out for most waste items.

The factors that have the largest impact on the 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 (measured in grams of Pu240e) is 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.

To quantify the A-term of the LLD, eight replicate measurements of a full (0% void), blank matrix (no Pu loaded) may be collected for SWB surrogate matrices. The replicates should comprise a no-source assay measurement with a surrogate matrix box paired with an empty chamber background measurement. For

each matrix, LLD_A (measured in grams of Pu240e) can be determined by applying group statistics to a sample of repeat assays carried out on blank waste matrices.

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

$$MDC = 10^6 \frac{Act_{spec}\sqrt{LLD_A^{\,2} + LLD_F^{\,2}}}{m} \qquad \text{(Eq. 5)}$$

where Act_{spec} is the specific TRU alpha activity of the isotope or mixture of interest (Ci/g), m is the container's net weight (kg).

The relationship between LLD (in terms of g WG Pu) and MDC (nCi/g for WG Pu) is plotted in Figure 3. 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 Figure 3 shows LLD and MDC calculated at 6% Pu240/Pu (WG) isotopics. For 12% and 18% Pu-240/Pu isotopics, both the LLD and the MDC are lower than for the illustrated 6% Pu-240/Pu (WG) case, for the entire net mass range shown in Figure 3.

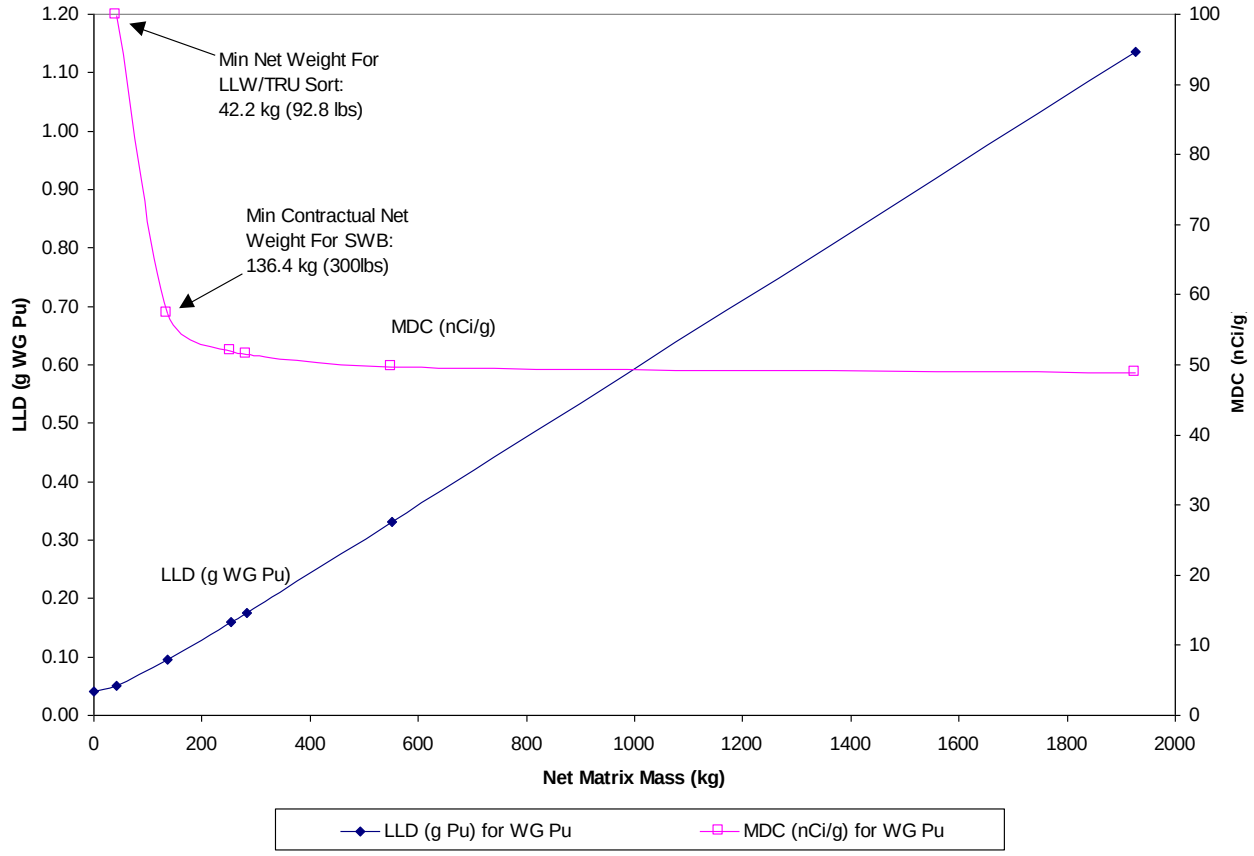

Fig. 3. Neutron LLD and MDC plot against net matrix mass

SUMMARY

The results of this study demonstrate the versatility and flexibility of the SuperHENC platform in characterizing a diverse range of matrix materials. Waste forms such as combustibles, plastics, metals, compacted pucks, and sludge have been successfully measured to date.

It has been demonstrated that the system meets all applicable regulatory performance objectives at several DOE sites (RFETS, Hanford and INL) and provides for TRU/LLW sorting of un-segregated debris waste with diverse isotopic mixtures.

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