

# QUALIFICATION OF BOXED WASTE NON-DESTRUCTIVE ASSAY SYSTEMS FOR MATERIALS CONTROL AND ACCOUNTABILITY MEASUREMENTS AT RFETS

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## ABSTRACT

As the Deactivation and Decommissioning (D&D) programs, at sites such as Rocky Flats Environmental Technology Site (RFETS) progresses, the focus is moving from assay of 55 gallon (208 liter) drums towards larger systems capable of assaying boxed waste forms. In the absence of specific regulatory performance criteria for boxes, non-destructive assay (NDA) data quality objectives (DQOs) are essentially independent of container size. Thus the box assay systems are required to meet DQOs originally promulgated for 55-gallon drum assays. Two new box assay systems have been deployed at RFETS. The Super High Efficiency Neutron Coincidence Counting System (SuperHENC) was designed and fabricated as a collaborative effort between RFETS, Los Alamos National Laboratory (LANL) and BNFL Instruments Inc (BII). The Multi-Purpose Crate Counter (MPCC) designed and built by BII is based on the Imaging Passive Active Neutron IPAN™ technology. The latter was designed to provide diverse capacity for assay of Standard Waste Boxes (SWBs) and for the larger B25 LLW crates. Both systems have been calibrated and validated in accordance with the requirements for RFETS Material Control and Accountability (MC&A) and WIPP disposal. The systems have achieved the matrix specific qualification criteria for precision and accuracy for safeguards termination limit assays over their calibrated range, including the new RFETS mixed D&D materials waste streams. These systems were the first, and at this time the only, waste box assay systems that have achieved both MC&A qualification and WIPP certification. They are now fully operational at RFETS and continue to support the site closure mission. This represents a significant milestone in both waste management at RFETS and in the U.S. Department of Energy's D&D program as a whole. This paper discusses the calibration and validation process undertaken to achieve the goal of MC&A qualification for the systems within the framework of the site's safeguards requirements.

## INTRODUCTION

Kaiser Hill with the support of BNFL Instruments (BII) and Los Alamos National Laboratory (LANL) has recently deployed two new crate assay systems at RFETS:

- The Super High Efficiency Neutron Coincidence Counting System (SuperHENC) a collaborative effort between RFETS, LANL and BII. This counter combines Neutron Coincidence Counting with Gamma Energy Analysis to assay SWBs with a sensitivity that allows for sorting at less than 100 nCi/g (3700 Bq/g). The system has been in operation since early 2001.
- The BII Multi-Purpose Crate Counter (MPCC) based on the Imaging Passive Active Neutron / Gamma Energy Analysis (IPAN™/GEA) technology. This crate counter provides diverse capacity assay of SWBs and larger LLW crates. The MPCC has operated since early 2002.

These assay systems provide the capacity to meet the assay requirements associated with the Deactivation and Decommissioning (D&D) at RFETS. This paper summarizes the MC&A qualification of these systems. This effort represents a significant milestone in D&D activities because (i) Performance criteria were achieved based on criteria originally promulgated for 55 gallon drum assay (ii) For the first time, qualification was achieved for the RFETS new mixed materials waste streams.

## MC&A PERFORMANCE REQUIREMENTS

Both the MPCC and SuperHENC are required to identify and analyze the presence of plutonium, uranium, americium and other elements of interest in varying mass ratio concentrations in low, medium and high density waste matrices contained in various container sizes.

The RFETS MC&A requirements for precision and accuracy [2] are summarized in Table 1. The accuracy requirement is particularly challenging for crate assay systems because allowance must be made for the measured precision i.e. the 95% confidence interval (derived from 2 tailed Student's T value) for the mean percent recovery (%R) must fall in the range given in Table 1

RANGE OF PU AND U (g)	PRECISION <sup>a</sup> (%RSD)	ACCURACY <sup>b</sup>
≤1.0	≤ 50	50 to 150
>1.0 to ≤10	≤ 25	75 to 125
>10.0	≤ 10	85 to 115

<sup>a</sup> Ratio of standard deviation in measured values of the known value, expressed as a percent

<sup>b</sup> 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.

**Table 1. MC&A Quality Assurance Objectives for Radioassay**

## MEASUREMENT TECHNIQUES

SuperHENC measures the <sup>240</sup>Pu<sub>eff</sub> content using passive neutron time-correlation counting and calculates the total plutonium content using Acceptable Knowledge (AK) for the plutonium isotopic mass fractions (<sup>238</sup>Pu, <sup>240</sup>Pu and <sup>242</sup>Pu are the only significant spontaneous fission sources in the RFETS weapons grade Pu waste stream). The system is operated by a custom tailored version of the LANL NCC software [2]. A <sup>252</sup>Cf add-a-source (AAS) is used for matrix correction [3]. Integration of neutron data with the gamma sub-system is performed with BNFL Instruments' SuperHENC Gamma Energy Analysis System (SGEAS) software. This uses the BII Efficiency Times Attenuation (ETA) technique, which is optimized for the sparse counting signal characteristic of large packages and dense matrices. All WIPP reportable quantities such as TRU alpha activity concentration, decay heat and <sup>239</sup>Pu fissile gram equivalent content are calculated by SGEAS.

The MPCC uses the IPAN method to quantify  $^{239}\text{Pu}_{\text{eff}}$  and  $^{240}\text{Pu}_{\text{eff}}$  in the respective active and passive modes of operation. The threshold for active/passive neutron mode selection has been set at  $0.6\text{g }^{240}\text{Pu}_{\text{eff}}$  (i.e.  $\sim 10\text{g WG Pu}$ ). When the measured  $^{240}\text{Pu}_{\text{eff}}$  passive mass exceeds this level, the passive result is used to determine the total plutonium. Below this level the active mode is used. Matrix sensing is performed with specialized neutron detectors known as ‘flux monitors’. The MPCC also includes a GEA gamma sub-system for assay of individual nuclides using the ETA technique.

### COMMISSIONING

The SuperHENC was installed at the site in December 2000. This is an entirely self-contained mobile system (figure 1) mounted on a commercial 48 ft. (14 m) long “low boy” trailer. The system was commissioned in January 2001.



(a) System installed at RFETS site



(b) Aft view

**Figure 1. SuperHENC/ SGEAS trailer**

The MPCC was delivered to the site on October 1999. Figure 2 shows the system at the factory. For site installation at RFETS, the MPCC was installed into a mobile enclosure. MPCC commissioning was performed in March 2002.



**Figure 2. The MPCC at factory prior to installation in mobile enclosure.**

## **CALIBRATION**

The SuperHENC measures the  $^{240}\text{Pu}_{\text{eff}}$  content using standard passive neutron doubles analysis. The calibration is based on an empty SWB doubles calibration curve with sample specific corrections applied for background and matrix effects.

The calibration was conducted in several steps. These steps included (i) constructing a Monte Carlo N-Particle (MCNP) model for the system, (ii) mapping chamber response with a neutron source, (iii) obtaining calibration measurements and establishing the coincidence calibration curve, (iv) establishing of the AAS correction factor calibration, (v) implementing background reduction techniques for samples with high backgrounds and low signals and (vi) validation of the calibration on different Pu standards at the final location (RFETS).

Calibration measurements using the MPCC were performed in 1999 at the Arkansas factory and in 2002 at RFETS. A set of calibration measurements was acquired using  $^{252}\text{Cf}$  and depleted uranium surrogate matrices. Measurements were performed for various surrogate matrices with the source positioned at 36 reference points in each crate (fewer positions are required in the empty crate). At the end of the calibration process, the passive background rates were corrected to reflect the background count rates at RFETS.

A technique for matrix selection in both active and passive mode making good use of the above calibration libraries has been developed for IPAN™ crate systems such as the MPCC. Data acquired from the flux monitors are used to determine measured indices (known as ABSMOD and MOD). These are used to select the appropriate calibration matrix for each measurement. With this method, the calibration selection is predicated upon by the waste's neutronic properties and is not limited to the specific materials used in the calibration (e.g metals or plastics).

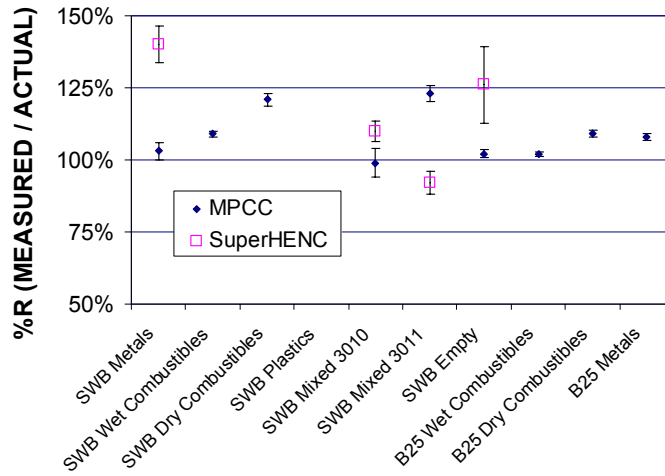
## **VALIDATION**

In order to demonstrate compliance with the MC&A performance objectives, validation measurements were collected on a variety of surrogate matrices that are representative of the waste stream at RFETS. Six replicates on three Pu loadings (1, 10, and 320 g) were collected. Sources were located in the approximate volume average position.

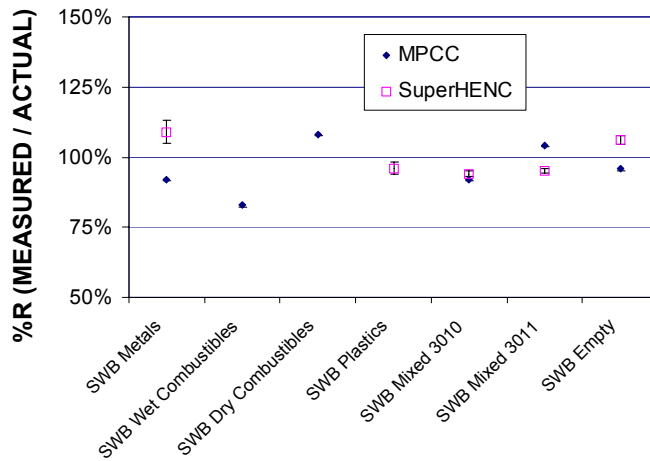
The surrogate matrices comprise SWBs loaded with a modular matrix cube design representative of RFETS materials such as mixed metals, dry combustibles and plastics. Traditionally RFETS has segregated its waste streams into well defined "item description codes" (IDCs) such as metals, dry combustibles and plastics. More recently, mixed matrix IDCs have been defined: (i) inorganic matrices with less than 10% by weight of organics (IDC 3010), (ii) inorganic matrices with greater than 10% by weight of organics (IDC 3011).

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

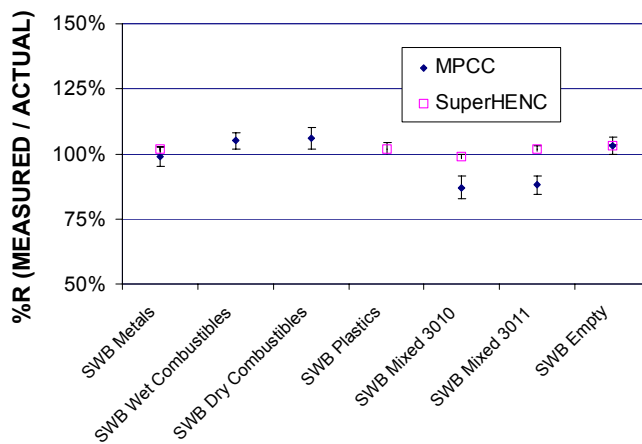
Charts 1 through 3 give a summary of the SuperHENC and MPCC validation measurements taken at RFETS [4] on independent Pu standards. None of these test standards had been used in the calibration of the systems. The charts show the Percent Recovery (%R) defined as the ratio of mean measured value to the actual certified Pu value expressed as a percent. The error bar on the charts indicates the percent relative standard deviation in the replicate assays. Data was collected with the standard operating procedure and assay parameters used in routine operations. All of the measurements shown here passed the applicable MC&A data quality objectives.



**Chart 1. Validation with 1g Pu**



**Chart 2. Validation with 10g Pu**



**Chart 3. Validation with 320g Pu**

## LOWER LIMIT OF DETECTION

Lower limits of detection (LLD) have been determined for both systems over their respective calibrated ranges. LLD 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. LLD is determined by a statistical analysis of replicate assays of blank surrogate matrices (i.e. containing no added activity). The LLD must account for interferences from different matrix conditions and the on-site radiation background (dominated by the cosmic ray background).

The LLDs are summarized in Table 2 for various surrogate matrices for the SuperHENC and MPCC [4]. All LLDs are comfortably below the lower limit required for MC&A performance objective demonstration of 1g Pu. These values may be converted to minimum detectable concentration (MDC) by converting to alpha activity for Weapons Grade Pu and dividing by net matrix weight. The MDCs are all less than 100 nCi/g (3700 Bq/g), enabling these system to perform TRU/LLW sorting for WIPP disposal.

Matrix (IDC)	Matrix weight (kg)	SuperHENC LLD (g WG Pu)	MPCC LLD (g WG Pu)
Empty (000)	0	0.113	0.018
Metals (480)	599	0.193	0.029
Dry Combustibles (330)	300	0.183	0.048
Mixed (3010)	535	0.219	0.108
Mixed (3011)	449	0.159	0.083

**Table 2. Detection Limit Summary**

## OPERATIONAL SUMMARY

The combined SuperHENC/SGEAS system has been in full-time operation, measuring SWB waste at RFETS since early 2001. The MPCC has been in operation since early 2002. Both systems have achieved WIPP certification. These are the first, and at this time the only, waste crate assay systems to achieve such status in the DOE complex.

## CONCLUSIONS

The SuperHENC/SGEAS and the MPCC IPAN™/GEA assay systems are now qualified for MC&A measurements, certified for WIPP measurements, and are performing waste box assays with a demonstrated TRU/LLW sorting capability at the 100nCi/g (3700 Bq/g) level. Qualification of the new mixed-matrix waste streams crucial to the successful D&D of RFETS was demonstrated.

The SuperHENC developed by LANL, BII and RFETS, has demonstrated excellent performance results for SWBs over a diverse calibration range. The MPCC supplied by BII, vastly expands the site crate capability as it can accommodate waste boxes up to 11,000 pounds and can measure both SWBs and B25s within the calibrated range.

Both box assay systems have established significant milestones in achieving MC&A certification, especially given that they met the data quality objectives that were originally developed for 55 gallon (208 liter) drums.

## **REFERENCES**

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- [4] Simpson A.P, et al, Operational and Regulatory Performance of Waste Crate Assay Systems at RFETS, Waste Management 03, Tucson, Arizona, February 2003.