Managing Uncertainty: Using the High Efficiency Neutron Counter to Efficiently Characterize Downblended Plutonium at Savannah River Site K-Area – 24273

Adam Gallegos[1], Joe Harvill[2], Daniel Remington[1] Sean Stanfield[1],
William Mussman[1], Timothy Aucott[3], William Searcy[1], Kevin Haar[2], Robert Semon[2],
Timothy Carlton[1], Jenna Lente[2]
1 Mirion Technologies
2 Salado Isolation Mining Contractors
3 Savannah River Nuclear Site

ABSTRACT

In August of 2021, the Waste Isolation Pilot Plant (WIPP) Central Characterization Program (CCP) and Mirion Technologies deployed 2 High Efficiency Neutron Counters (HENCs) at the Department of Energy Savannah River Site (DOE-SRS) K-area Facility to characterize downblended surplus plutonium in the state of South Carolina. The characterization program at SRS K-area has been successful due to the technological forethought in the development of a precise waste product, container packaging selection and nondestructive assay (NDA) calibration accuracy and total measurement uncertainty (TMU). These features all play pivotal roles in maximizing waste characterization efficiency.

To date, the HENC NDA units have characterized approximately 1200 Criticality-Control Overpacks (CCOs) at SRS K-area. The WIPP Transuranic Waste Acceptance Criteria limits each CCO container to a total of 380g Pu-239 Fissile Gram Equivalent (FGE), including 2 times the calculated FGE uncertainty. The development of the CCO was intended to increase the radiological loading limit of waste material by providing more radiation shielding and enhanced criticality protection. The CCO containment selection increased the total FGE packaging efficiency by 90%, when compared to a standard 55-gallon drum (200 FGE limit).

Once the designated containment was selected, adequate CCO calibration standards were developed to create an empirically derived passive neutron calibration curve and confirm an In-Situ Object Counting System (ISOCS) calculated HENC calibration. Using well characterized and quantified CCO calibration standards, the HENC system measurement calibration confirmation measurements results performed successfully, measuring within ±3% (neutron) and ±4% (gamma) respectively of the known Pu loading. The development and performance of the HENC neutron and gamma calibration has informed the waste loading operations by providing a baseline performance metric for relatively pure downblended Pu waste. Historical use of the total measurement uncertainty calculations have been challenging due to variable waste characteristics associated with a particular waste stream: matrix material non-uniformity, variations in waste density, measured isotopic uncertainty and low-z impurities. When each characteristic is applied to a further variable waste stream, each uncertainty category can contribute to large individual segments of the overall applied uncertainty to the total FGE calculation. Because the downblended waste material from SRS is pedigreed and highly engineered, the individual uncertainty contributions can be efficiently controlled. Through the efforts of DOE-SRS, CCP and Mirion, the total measurement uncertainty has been improved through modeling efforts of the waste material, packaging and impurities—which has allowed for a more precise TMU calculation and hence a more efficient packaging configuration.
Future waste processing facilities can use the SRS K-area waste characterization project as a case study in how technical planning and cooperation with the private sector can increase worker safety, equipment efficiency and cost savings. The catalogue of nondestructive assay tools and techniques combined with extensive knowledge of programmatic requirements should therefore be essential when developing or improving a waste characterization program at nuclear facilities.

Celebrating 25 years of operations at the speed of safety, WIPP continues to be the cornerstone for DOE's nuclear waste cleanup efforts, removing legacy TRU waste at 22 sites while reducing risk for Americans living near these sites, now and into the future.

INTRODUCTION

The SRS K-area facility is working to characterize and dispose of 34 metric tons of surplus plutonium as part of the Department of Energy and National Nuclear Safety Administration (NNSA) commitment to the state of South Carolina. The downblended surplus plutonium is packaged in CCO containers, characterized using the HENC NDA system and approved for shipment following the DOE Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant (WIPP/WAC). By selecting the HENC NDA instrument, CCP NDA personnel have the flexibility to measure and analyze the packaged nuclear material by quantitative gamma analysis, passive neutron coincidence counting or using a combination of both modalities. At present, 5 total HENC instruments have been deployed for CCP operations: 3 units operating at Los Alamos National Laboratory (LANL) and 2 units operating at SRS.
The HENC is configured to directly quantify ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{237}Np, ^{233}U, ^{235}U, ^{238}U, ^{241}Am, ^{137}Cs, and other radionuclides by gamma spectroscopy [1]. Other gamma-emitting radionuclides are also being determined by direct measurement. A single Broad Energy Germanium (BEGe) gamma ray detector acquires spectra between approximately 86 keV and 1500keV for direct quantification of those radionuclides, as well as for plutonium isotopic analysis. The Pu isotopic ratios are determined using the MultiGroup Analysis (MGA) or Fixed-energy Response-function Analysis with Multiple-efficiencies (FRAM) software, or Acceptable Knowledge (AK).

Fig. 1: High Efficiency Neutron Counter NDA System

Passive neutron measurements use ^{3}He proportional counters for neutron detection and both coincidence and multiplicity signal analysis to directly measure the spontaneous fission content in waste drums. The detector bank assembly has a total of 113 ^{3}He detector tubes, 1-inch in diameter with active lengths ranging from 25 inches to 45 inches [1]. The HENC chamber is enclosed by high-density polyethylene (HDPE) to moderate source neutrons for more efficient passive neutron detection and shielding of background neutron interferences (Fig. 1). The system is operated using Mirion Technologies' Genie 2000 and NDA 2000 software platforms.

In 2021, CCP NDA personnel calibrated the HENC gamma and neutron modalities, and the calibrations were confirmed through the CCP calibration confirmation process, as outlined by the DOE WIPP/WAC. The HENC calibration confirmation results are documented in the CCP report listed in Reference 1. Along with the HENC system calibration and confirmation, the WIPP/WAC requirements also state a total measurement uncertainty estimate must be documented and technically justified for each certified NDA assay system [2].

CCP TMU technical analysis and reporting is documented across two documents. The HENC passive neutron coincidence uncertainty was developed by SRS, LANL and Pacific Northwest National Laboratory (PNNL) personnel and reported in *HENC Modeling Measurement Uncertainties* PNNL-31054. The HENC quantitative gamma analysis TMU is documented in *Savannah River Site High Efficiency Neutron Counter Total Measurement Uncertainty Report* CI-HENC4-HENC5-TMU-1001. The primary objective of each TMU analysis is to define each uncertainty contributor and factor the measurement impact into the initial calibration, such that the contributed uncertainty could be minimized. To this end, the SRS downblended Pu material has relatively well understood measurement dynamics and TMU contribution.

Due to the large volume of waste material in this waste stream, SRS and CCP NDA groups are motivated to efficiently measure and dispose of the surplus plutonium. Therefore, much technical forethought has been placed into all aspects of the HENC NDA operations and analysis process. The primary areas of focus of this discussion are the HENC NDA measurement accuracy and the CCP FGE disposal limit as impacted by the TMU.

DESCRIPTION

Waste Package Containment

Waste containers packaged across the DOE complex typically contain highly variable material types and densities, ranging scrap metal, plastic tubing, electrical devices, rubber gloves taken from decontamination and decommissioning activities [3]. Because of this diversity of constituents, the NDA system total measurement uncertainty budgets can be equally variable due to container measurement effects such as shielding, matrix-material non-uniformity, density variation, chemical composition etc. However, one of the primary benefits of measuring downblended CCOs from SRS K-area is the material constituents and containment is intended to perform as an engineered product, thus minimizing the TMU contributions of those previously mentioned. Since the goal is always to efficiently and safely package, characterize and ship compliant waste containers to WIPP, the SRS K-area CCO is an optimal configuration for generating a computational model that is representative of the measurement dynamics of the system.

The CCO consists of a criticality control container (CCC) positioned by plywood upper and lower dunnage assemblies within a 55-gallon drum (Fig.2) [4]. The CCC body is constructed of stainless steel 6-inch Class 150 standard blind and slip flanges and Schedule 40 pipe. The manufactured waste form is loaded into the internal CCC. No waste material is directly loaded into the CCO.

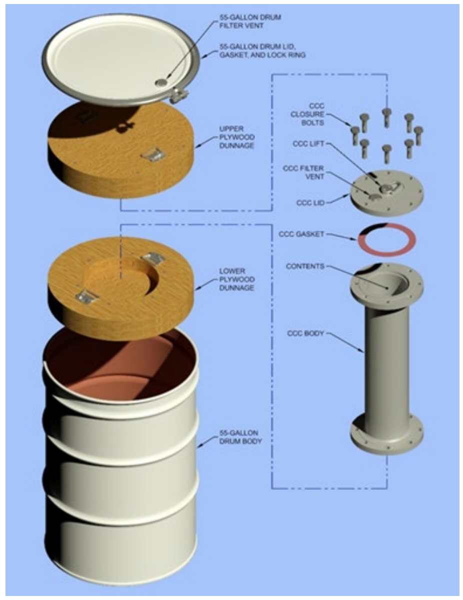

Fig. 2: Expanded View Criticality Control Overpack [4]

Quantitative Gamma and Passive Neutron TMU Estimates

Because each waste package is comprised of very consistent materials and known Plutonium content, the quantitative gamma analysis functions of Mirion Technologies ISOCS software were ideal for generating a TMU calculation. In addition to calculating efficiency calibrations for custom or complex geometries, the ISOCS software has the ISOCS Uncertainty Estimator (IUE) that is utilized to perform a variety of total measurement uncertainty calculations for a respective ISOCS model (Fig. 3).

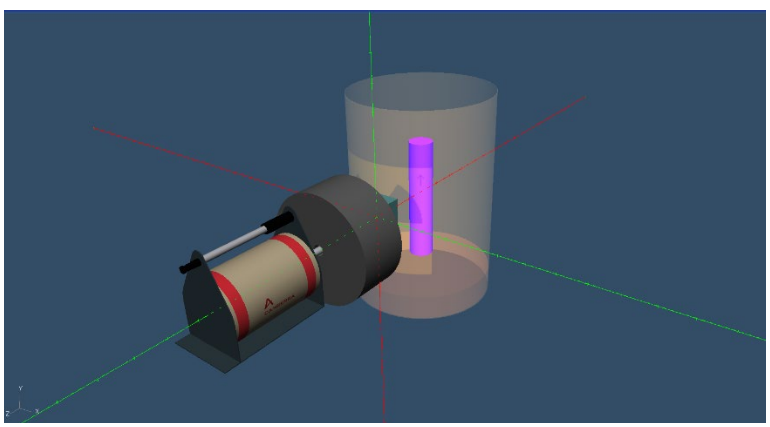

Fig. 3: ISOCS model of the CCO waste container used to develop the gamma calibration curve and TMU estimate.

ISOCS allows the user to vary each of the model parameters in order to generate the TMU relative minimum (R_{min}) and maximum (R_{max}) values, system sensitivities, assay uncertainties, and optimization values for a given ISOCS model of interest. This software package was utilized to generate the TMU values, listed in **Table 1**.

Table 1. Quantitative Gamma Analysis Total Measurement Uncertainty Budget [5]

Uncertainty	% at 1-σ
Counting Statistics	2.0
Calibration	7.2
Matrix Material Non-Uniformity	2.0
Variations in Container Density	8.0
Total TMU Budget	**11.1**

The TMU budget of 11.1% at 1σ represents a significantly narrower uncertainty range compared to the standard direct-loaded Pipe-Overpack Container (POC) for debris waste, which has an uncertainty of 12.2% at 1σ. [6]

The passive neutron HENC modeling and TMU analysis performed by SRS, LANL, and PNNL likewise resulted in an improved uncertainty budget. One of the primary contributors to SRS CCO uncertainties are neutrons created by alpha-particle-generated neutrons from interaction with impurities in the material such as beryllium, oxygen, and fluorine [7]. Specifically, the level of impurities is α, the ratio of the neutrons generated from (α, n) reactions to the neutrons from spontaneous fission. The impurity ratio is expected to result in a coincident neutron measurement bias. Thus, the downblended plutonium CCOs have been categorized into families; pure material, impure material, very impure material and mixed uranium/plutonium. Each family is further divided into subfamilies, based on 240Pu/239Pu ratio for pure materials or on the nature of the impurities for the other families. This categorization is outlined in Table 2 below. The measurement subfamilies used and applicable for this discussion are 1A (Aries) and 2D (Pyro).

Table 2. Plutonium measurement families and subfamilies [7]

1: Pure Material (α < 3, U < Pu)	2: Impure Material ($\alpha \leq 10$)	3: Very Impure Material ($\alpha \geq 10$)	4: Mixed U/Pu (α < 3, U ≥ Pu)
1A: ARIES Material	2A: Low Be	3A: High Be	4A: High U-235
1B: Weapons Grade	2B: Low Be/Fl	3B: High Be/Fl	4B: High U-238
1C: Fuel Grade	2C: Low Fl	3C: High Fl	4C: Other U/Pu
1D: Reactor Grade	2D: Pyro	3D: High Mg	4D: Very High U
	2E: Other Low Impurities	3E: Other High Impurities	

Based on the HENC system detection modeling, and TMU contributors such as predicted measurement bias, isotopic and chemical composition, background, source and matrix distributions, a TMU budget for each subfamily was calculated such that each CCO could have an individual budget relative to its contents. Each subfamily TMU results are stated in Table 3.

Table 3. Uncertainty Budget for Total Measurement Uncertainty of FGE [7]

Sub-family	α range	Counting (%)	Chemical Form (%)	Nominal TMU 1σ (%)
1A	1-3	1.1	1.4	4.5
1B	1-3	1.0	1.0	4.4
1C	1-3	2.0	1.9	5.0
1D	1-3	3.7	2.8	6.2
2A	1-10	1.4	6.8	8.1
2B	1-10	1.4	6.8	8.1
2C	3-10	1.4	6.8	8.0
2D	3-10	1.4	6.7	8.0
2E	3-10	1.4	6.7	8.0
3A	10-100	3.6	59.2	59.4
3B	10-50	3.0	34.2	34.5
3C	10-80	4.0	51.1	51.5
3D	10-20	2.0	12.2	13.0
3E	10-20	2.0	14.0	14.8

For families 1 and 2, the modeled results show a nominal TMU range for CCOs at 1σ of 4.5% - 8.1% [7]. Again, the SRS HENC TMU result is very narrow as compared to the POC debris waste neutron uncertainty budget of 14.2% at 1σ [6].

The measurement modality (gamma or neutron) TMU estimate is applied on a container-by-container basis, calculated within Mirion Technologies software NDA 2000. Minor variances can be observed across a population and measurement anomalies are expected. The final HENC TMU budget is factored into the reported CCO WIPP/WAC characterization results for all containers. Because the SRS loading procedure is intended to package efficiently, one of the primary goals is to ensure each CCO does not exceed the WIPP FGE disposal limit described in the next section.

CCP Measurement Accuracy and CCO ^{239}Pu FGE Limit

For a CCO container to be accepted into WIPP, the sum of ^{239}Pu FGE plus two times its associated TMU, expressed in terms of one standard deviation, shall not exceed 380[2].

$$^{239}\text{Pu FGE}+2\sigma \leq \text{FGE}_{limit}\ 380 \qquad \text{[Eq. 1]}$$

Each CCO container generated at SRS is relatively well characterized, as isotopics and total masses have been consistently tracked by SRS Nuclear Measurement Control and Accountability. From combined knowledge of the expected Pu mass, isotopic distribution and WIPP loading limit, site packaging operations can efficiently package containers with the expected measurement results remaining compliant. However, because the impurity impact on a container-by-container basis is unknown, there remains likelihood of measurement anomalies resulting in unfavorable results.

This level of uncertainty has prompted the CCP NDA group to continually monitor and track measurement results for each subfamily, to ensure the expected outcomes are reached. One continually tracked metric is the percent recovery (%R) of each downblended CCO.

$$\%R = \frac{Measured\ Source\ Activity}{Known\ Source\ Activity} * 100\% \qquad \text{[Eq. 2]}$$

The CCP historically uses the accuracy test of %R in various calibration testing procedures, thus it was appropriate to monitor the system accuracy for each of the CCO subfamilies. Tracking %R across subfamilies will assist CCP and SRS personnel in guiding the packaging and analysis process for each subfamily and ensure technical problems can be addressed when encountered. The primary benefit of having a well characterized material tracking effort prior to the certified measurement is narrowing the unknown factors that can impact the measurement results. At times, the CCP NDA measurement group has consulted with SRS engineering to resolve challenges such as isotopic uncertainty and incomplete historical data. Subsequently, these problems are often quickly resolved as issues that can be effectively mitigated.

Below are the results for all current 1A and 2D subfamily Plutonium mass %R. The number of containers measured to date are 224 and 158, respectively (Fig.4 & Fig.5).

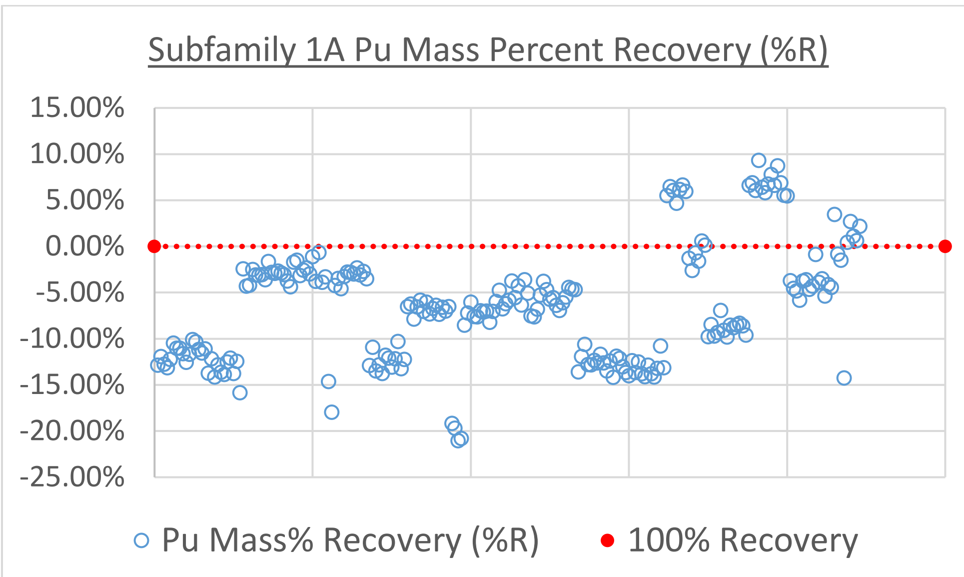

Fig. 4: Subfamily 1A Total Plutonium Mass %R

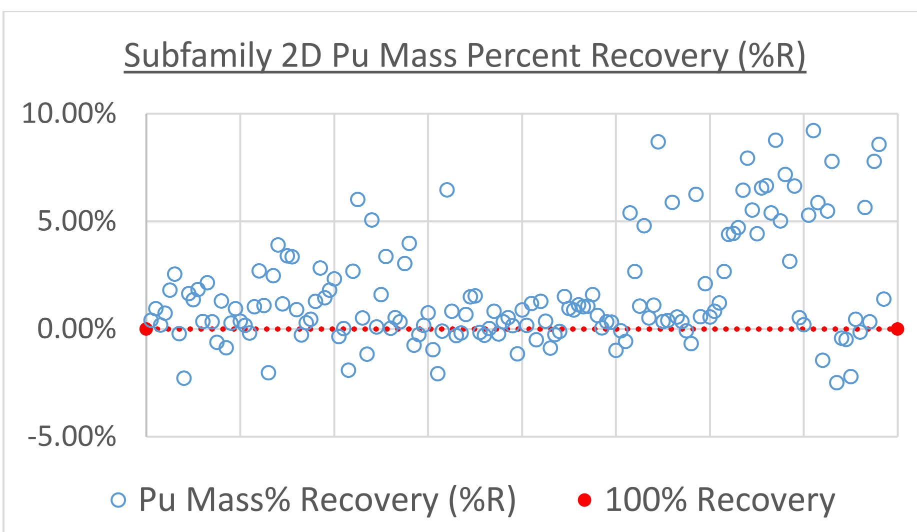

Fig. 5: Subfamily 2D Total Plutonium Mass %R

Below are the measurement results for Subfamily 1A and 2D WIPP/WAC CCO FGE disposal limit (Fig.6 & Fig.7).

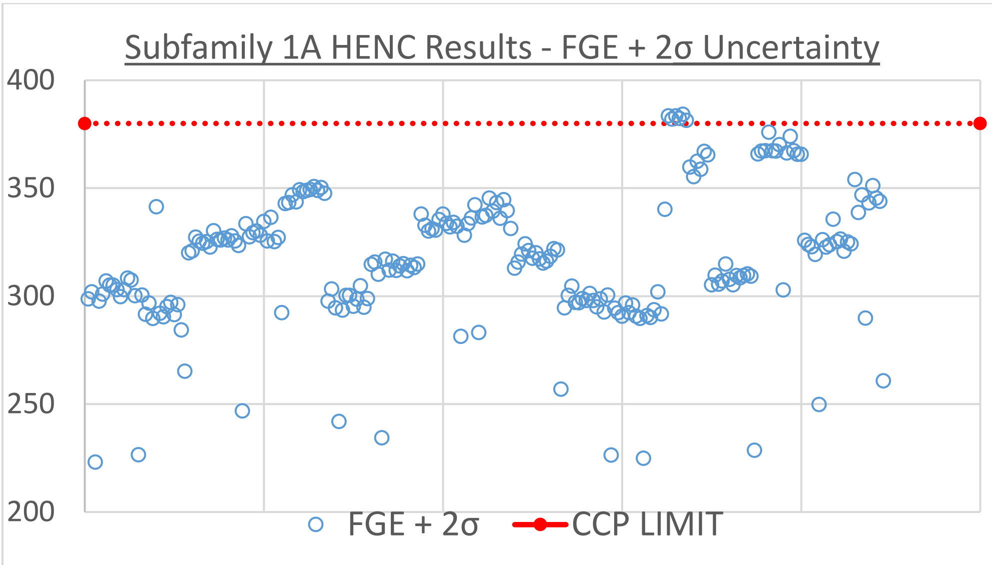

Fig. 6: Subfamily 1A ^{239}Pu FGE + 2 σ HENC Measurement Results

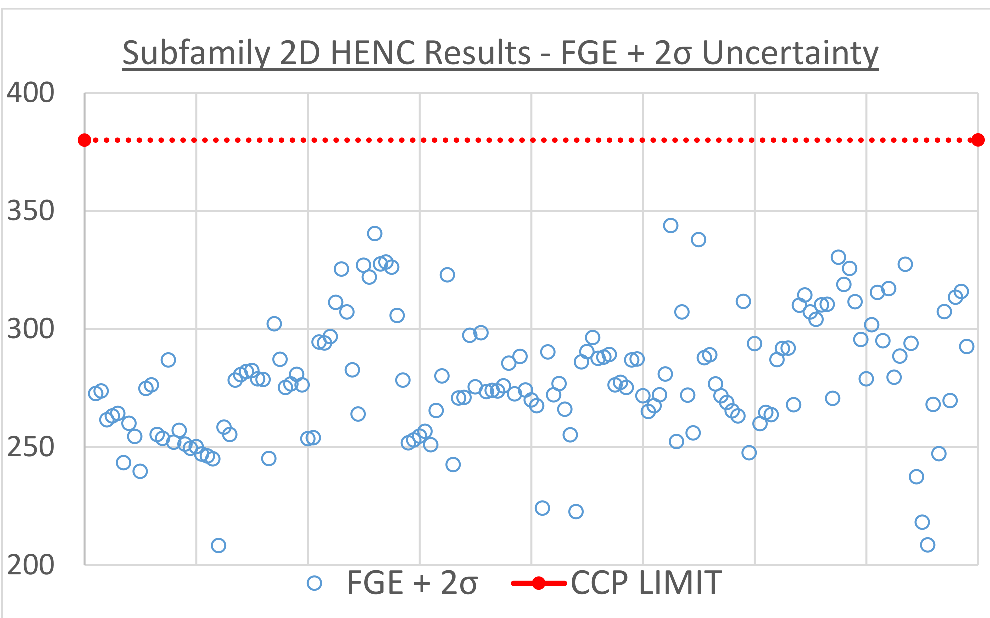

Fig. 7: Subfamily 2D ^{239}Pu FGE + 2 σ HENC Measurement Results

DISCUSSION

Over the course of evaluating Subfamily 1A, a slightly negative bias was encountered among the initial measurement population. During the subsequent weeks, historical data was scrutinized and measurement accuracy of isotopics and uncertainties were reviewed against calorimetry and destructive analysis data to find and evaluate discrepancies. Over time, the %R improved as the preliminary characterization was improved. Likewise, Subfamily 2D has shown very positive initial results. Additionally, the most recent measurement data are being monitored to ensure an upward bias trend across downblended containers does not worsen.

Regarding the WIPP/WAC FGE disposal limit, the majority of containers remain comfortably below the 380 threshold. Results exceeding the FGE threshold were quickly identified, investigated and resolved by working in concert with SRS engineering. Because the 2D measurement performance has shown to have a relatively low bias effect on the final %R result, there may be further opportunities to increase loading per container.

CONCLUSIONS

The combined efforts of the CCP and SRS to optimize the K-area cradle-to-grave characterization program has shown immediate success as a technically planned operation. The foresight of targeting the measurement TMU as a method of improved packaging waste efficiency is proving to be an effective method of informed practice. Because NDA analysis across the DOE complex has been extremely dynamic, the TMU analysis was always extremely conservative and thus negatively impacting the ability to ship efficiently. Admittedly, the CCO is an optimized measurement condition for waste disposal. However, DOE facilities can glean insight from the SRS K-area dilute and dispose operation by reducing the impact of known challenges through technical means. For example, the SRS engineering team is using the resulting CCP measurement data to locate areas of improvement. Early candidates for further scrutiny are isotopic accuracy and uncertainty and combining of subfamilies. Likewise, preliminary measurements of very impure materials from Family 3 are being measured to observe the system behavior prior to committing to a loading strategy.

The decades of measurement experience from Mirion Technologies, SRS and CCP have converged to meet a difficult challenge for the DOE and NNSA. During the upcoming years, the NDA program will continue to fine-tune the analysis methodologies, review areas for TMU improvement and be ready to resolve technical problems.

REFERENCES

1. Savannah River Site HENC4 and HENC5 ISOCS Calibration Report CI-HENC4-HENC5-NDA-1000, Rev 0, 2021
2. Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant, DOE/WIPP-02-3122 Rev. 11, 2022
3. Walker, Connie, Evaluation of the Radiological Characterization of Savannah River Site Waste Stream SR-W027-221H-HET-C, Revision 3, 2020
4. Specification For Criticality Control Overpack Performance Demonstration Program Fabricated Steel Insert, Revision 1, October 2020, Nuclear Waste Partnership, Carlsbad, NM
5. Savannah River Site High Efficiency Neutron Counter Total Measurement Uncertainty Report, CI-HENC4-HENC5-TMU-1001, 2021

6. Total Measurement Uncertainty for the HENC #3 With Integral Gamma Spectrometer, CI-HENC3-TMU-101, Revision 0, 2014
7. Aucott, Timothy J., Geist, William H., Mendoza, Paul M., Moore, Michael E., Siciliano, Edward R., Warren, Glen A., HENC Modeling Measurement Uncertainties, PNNL-31054., 2021.