Novel Gamma Attenuation Methods Utilized for Waste Characterization of UO_2F_2-Bearing Materials at the Paducah GDP – 17327

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ABSTRACT

The Paducah Gaseous Diffusion Plant (PGDP) is currently under deactivation with demolition in the decades to follow. UO_2F_2-bearing waste is generated at PGDP by UF_6 hydrolyzing to materials such as pipes, valves, PPE, etc. when UF_6 is exposed to humid air. In addition, Al_20_3 and NaF trap materials contain significant amounts of Uranium. The UO_2F_2 deposits within the generated waste can be of sub-gram levels to several hundreds of localized grams and are much denser than the surrounding interfering matrix materials and may be of very complex geometric and chemical forms such as with the Al2O3 and NaF trap materials. The main objective is to characterize the fissile (U-235) content of the UO_2F_2-bearing waste so as to meet the Data Quality Objectives (DQO) for proper disposal. If a waste container cannot meet the DQO then it must be subjected to costly remediation, repackaging and re-characterization processes. We have developed, at PGDP, several novel model-based methods and techniques that vastly improve the standard gamma attenuation correction methods and do not require Working Reference Materials (WRM) for calibration. These new methods involve rotating and non-rotating 18.9 liter (5 U.S. gallon) waste containers with and without a transmission modality. A new model-based variant of differential attenuation has been developed that provides a high-quality self-absorption correction that does not require knowledge of the Uranium enrichment or isotopic ratios. For more challenging waste containers, a method using multi-view (non-rotating) 18.9 liter waste containers has been considered using a recently developed Advanced ISOCS© Uncertainty Estimator (AIUE) package. The new methods suffice to significantly improve accuracy and precision and to lower the Total Measurement Uncertainty (TMU) leading to satisfactory DQO's that were not previously achievable using the standard gamma attenuation correction methods. In this paper we present real 18.9 liter waste container and Working Reference Material (WRM) data subjected to the new methods and compared to standard methods depicting the significant improvement in the DQO that is realized.

INTRODUCTION

The Paducah facility has been under deactivation for the last two years and much effort has been in place to develop methods to characterize UO_2F_2 bearing waste for disposal purposes. A large portion of the UO_2F_2 waste is contained within 18.9 liter (5 U.S. gallons) pails with waste streams such as alumina and NaF trap mix as well as copper piping, inorganic sludges, PPE (Personal Protection Equipment) and

laboratory media. The waste streams can be quite non-uniform in Uranium loading with much clumping and experience considerable self-absorption negating the use of standard gamma attenuation correction techniques.

At the Paducah facility are two identical Q2 (Qualitative and Quantitative; Canberra model# WM-2110 [1]) units each recently configured with three HPGE (Canberra model# BEGE 3830) detectors. The Q2 instrument is designed for assaying 208 liter (55 U.S. gallons) drums; the introduction of a pedestal at the Paducah facility offers the ability to assay 18.9 liter (5 U.S. gallon) drums as shown in Fig 1. The detector spacing is optimized for 208 liter drums and cannot be altered but the pedestal height is adjustable and optimized for sensitivity with respect to the 18.9 liter pail. The data is acquired such that the detectors can be analyzed individually as well as the detector physical sum. The pails can be rotated continuously or rotated statically in 30 or 60 degree increments. In addition, a transmission source (Eu-152) can be placed on the Q2 inside wall so that it opposes the middle detector and transmits through the pail for various heights, using a lab jack, as shown in Fig. 2. With the current labjack, nine (9) vertical positions can be achieved.

Efficiencies

Gamma efficiencies were created in the standard way using ISOCS [2]. The VWA (Volume Weighted Average) multi-density curve technique was utilized for each of the waste streams. The recommended manufacturer maximum loading capacity of the pail is 22.7 kg (50 lbs.) so the efficiencies were generated for a range of densities between 0 and 3.0 g/cc.

Total Measurement Uncertainty (TMU)

The ISOCS Uncertainty Estimator (IUE) [3] was utilized to generate the multi-curve VWA (Volume Weighted Average) TMU (Total Measurement Uncertainty) budget for all the waste streams for a range of densities. The results are depicted in Table I for an average density of 0.5 g/cc. The largest TMU components are Rmin/Rmax, lumpy source and lumpy matrix. The Rmin/Rmax component represents point sources in the lowest and highest efficiencies relative to the VWA. The lumpy source component accounts for UO_2F_2 lumps ranging in various sizes and densities (up to 2.6 g/cc). The lumpy matrix component represents local deviations of the matrix with higher and lower (voids) matrix elements.

Non-Uniformity

A metals 18.9 liter (5 U.S. gallon) pail of true UO_2F_2 bearing waste containing copper pipes, valves, rags, PPE (Personal Protection Equipment) and Uranium precipitate was assayed with the lab jack at position 3 as shown in Fig 2. The pail was statically rotated to produce emission (186 keV) and transmission (344 keV) data for 0, 30, 60, 90, 120, 150 and 180 degrees. The results are shown in Fig. 3 where it is clear that there is high non-uniformity, within the pail, with respect to

the emission and transmission. Standard techniques of rotating the drum so as to produce VWA (Volume Weighted Average) results are generally successful but the analyst must include the possibility of non-VWA situations which increase the TMU (Total Measurement Uncertainty) budget quite significantly.

Self-Absorption

Considering the density of UO_2F_2 is approximately 2.6 g/cc it is observed that the relatively low-energy lines of U-235 suffer notable self-absorption, *i.e.*, the material is self-attenuation. This cannot be accounted for by using VWA (Volume Weighted Average) efficiency calibrations. The alumina trap mix consists of moistened alumina pellets where UF_6 gas has hydrolyzed to UO_2F_2 adsorbing as a spherical shell to the alumina pellet. The UO_2F_2 spherical shell can be quite thick as is evident in Fig.4 where the VWA determined mass range varies considerably over the U-235 energy lines. Simply taking the highest penetrating line of U-235 still produced results which are biased low.

Objective

The primary objective is to minimize the contribution of Rmin/Rmax, lumpy source and lumpy matrix TMU components to the TMU budget for a Q2 system. Reducing the overall TMU leads to achievable DQO's (Data Quality Objectives), *e.g.*, NRC 10CFR71.22 General License: Fissile Material, for U-235 mass shipping limits, which may produce results which otherwise could not lead to disposal. This requires the development of new methods which is the target of the remainder of this paper

Table I. 18.9 liter pail TMU budget (1-σ) for various waste streams at 0.5 g/cc.

TMU Type	186 keV 1-σ TMU (%) for 0.5 g/cc				
	DAW	Metals	NaF Trap	Alumina Trap	Inorganic Sludge
Rmin/Rmax	11.41%	12.20%	10.64%	11.86%	10.63%
Lumpy Source (LS)	11.62%	11.48%	9.12%	10.01%	18.79%
Lumpy Matrix (LM)	3.19%	22.47%	6.50%	11.81%	6.45%
Mixed Matrix (MM)	0.54%	0.54%	0.54%	0.54%	0.54%
Fill Height (FH)	1.59%	1.59%	1.59%	1.59%	1.59%
Drum Centering (DC)	1%	1%	1%	1%	1%
Mixed Enrichment (ME)	5%	5%	5%	5%	5%
Calibration	4%	4%	4%	4%	4%
Counting Statistics	2%	2%	2%	2%	2%
Total	18.01%	28.89%	16.95%	20.72%	23.59%

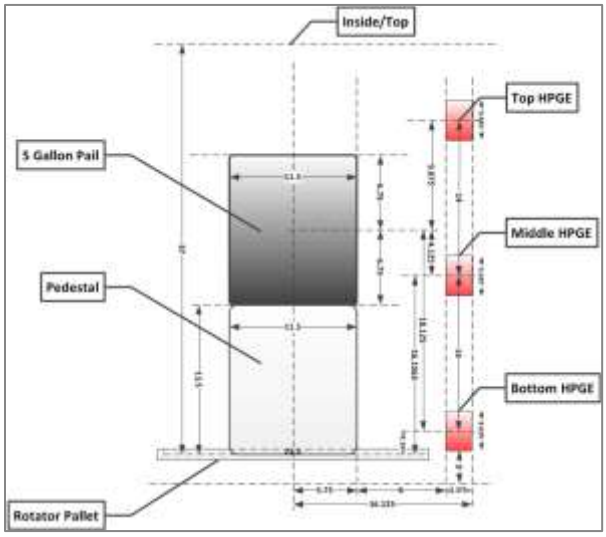

Fig. 1. The pedestal arrangement for 18.9 liter pails with the Q2 unit at the Paducah facility. Units are in inches.

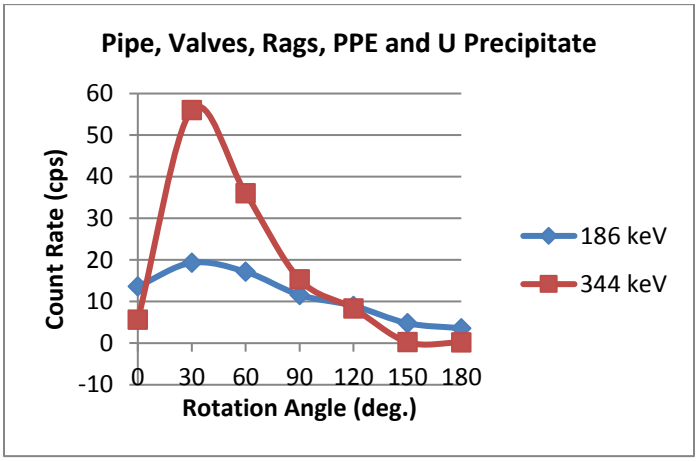

Fig 3. 18.9 liter pail metals waste drum showing the emission (186 keV) and transmission (344 keV) peaks as a function of rotational angle (degrees). The average density of the pail contents is 0.931 kg/liter.

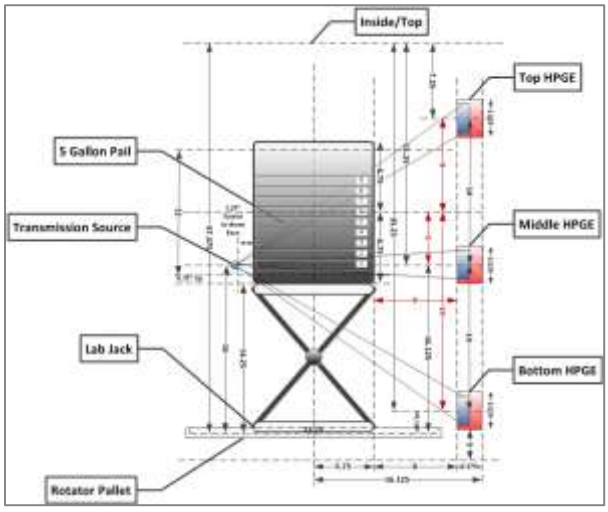

Fig. 2. The transmission arrangement, with adjustable lab jack, for 18.9 liter pails with the Q2 unit at the Paducah facility. Units are in inches.

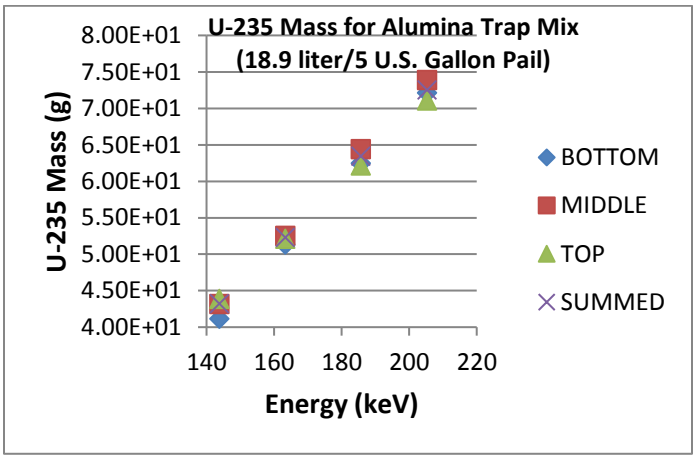

Fig. 4. Alumina trap mix waste pail showing the VWA, uncorrected, computed U-235 mass computed from each gamma line of U-235. The results are shown for the bottom, middle and top detectors as well as the detector sum.

METHODS

Several methods have been developed; all achievable with the current equipment at the Paducah site.

AIUE Method

The AIUE (Advanced ISOCS Uncertainty Estimator) acquisition and analysis method was developed at Canberra and is published elsewhere [4]. The implementation of the method for the FPDP Q2's consumes the ISOCS template utilized to generate the efficiencies as well as the measured isotopics, differential peak and multi-view (multi-count) as Figure of Merits (FOM's). The multi-count/view data is acquired for the geometrical layout shown in Fig. 5 for the 9 vertical positions depicted in Fig. 2. The data is also acquired for each detector. The acquisition data and parameters are configured into the AIUE program and the results are computed for each detector.

Self-Absorption Methods

Two methods have been developed; the first method, Self-Absorption Correction (SAC), requires Uranium isotopics or enrichment and that U-238 be in equilibrium with Pa-234m. The second method, differential attenuation, does not require isotopics nor that U-238 be in equilibrium with Pa-234m.

SAC

The NDA 2000 Self-Absorption Correction (SAC) engine is a standard infinite energy mass correction algorithm as defined in the literature [5]. The algorithm defines a mass curve which bends asymptotically towards E = infinity. The engine is designed for nuclides such as Pu-239 where gamma lines are fairly well separated as to allow confidence in fitting the mass curve – not so for U-235 since the lines are closely clustered and of relatively low-energy. If, however, we include, and relate, the measured U-238 1001 keV line peak area to U-235 via isotopics and associated declared constants, we can extend the standard SAC curve fitting to accommodate U-238 since the 1001 keV line will be included in the U-235 SAC fit.

The atomic mass ratio between U-235 and U-238 is defined as,

$$m_{235} = r \cdot m_{238} \qquad \text{Eq. 1}$$

Where the mass ratio, $\boldsymbol{r}$, is a variable which can either be declared (user entered) or measured with a suitable isotopics code. Utilizing the specific (mass) activity, $\boldsymbol{m_x = A_x \phi_x}$, we have,

$$A_{235} = r \cdot A_{238} \cdot \frac{\phi_{238}}{\phi_{235}} \qquad \text{Eq. 2}$$

Where ϕ_{238} = 2.9745 g/μCi and ϕ_{235} = 0.4626 g/μCi are accepted values at the Paducah site

The activity for the U-238 1001 keV line is defined as,

$$A_{238}|_{E=1001} = \frac{P_{238}}{b \cdot \varepsilon}\Big|_{E=1001} \quad \text{Eq. 3}$$

Where ε is the matrix corrected efficiency and b is the branching ratio (defined as b = 0.8370). The corresponding U-235 activity for the 1001 keV line would then be,

$$A_{235}|_{E=1001} = r \cdot \frac{P_{238}}{b \cdot \varepsilon} \cdot \frac{\phi_{238}}{\phi_{235}}\Big|_{E=1001} \quad \text{Eq. 4}$$

Substituting,

$$b' = b \cdot \frac{\phi_{235}/\phi_{238}}{r} \quad \text{Eq. 5}$$

We have,

$$A_{235}|_{E=1001} = \frac{P_{238}}{b' \cdot \varepsilon}\Big|_{E=1001} \quad \text{Eq. 6}$$

Where b' is the "effective", computed, branching ratio for U-235 with respect to the U-238 1001 keV peak area. The same principle can be extended to include other lines of U-238, e.g., 258 and the 766 keV if the lines are present in the spectrum. The engine (method) logic is outlined below.

- Determine the assay sequence isotopics preference, measured or declared.
- If preference is measured isotopics, attempt to get MGAU results – if no MGAU results, then revert to declared isotopics if available.
- Else, if preference is declared isotopics then attempt to get declared isotopics if available.
- If no isotopics available, or isotopics available but no U-235 and U-238 non-zero isotopics, then exit without error and without computing b'.
- Search the nuclide library associated with the assay sequence for the special 1001 keV line assigned to the ^{235}U nuclide designated for SAC.
- If the special 1001 keV line is not available then exit without error and without computing b'.
- Establish the mass ratio, r, from the measured isotopics.
- Compute b' and save to the nuclide library associated with the assay sequence.

The method is easily extended to the other lines of U-238 using the associated site accepted branching ratios.

Differential Attenuation

The differential attenuation method, as adopted in this paper, is a close cousin of the classical differential peak method [6] where peak pair ratios are exploited to determine the energy dependent self-attenuation correction factors (CF's). The main difference is that a differential peak calibration is not required for the differential attenuation method; instead only the VWA (Volume Weighted Average) ISOCS efficiencies, mass attenuation coefficients (MAC) for the Uranium bearing material and sufficient model representative of the geometry (spherical, rectangular or cylindrical).

The activity for each U-235 line is defined as,

$$A_{235} = \frac{P_i}{\epsilon_i \cdot b_i} \cdot CF_i \qquad \text{Eq. 7}$$

Where P_i is the gamma line peak rate, ϵ_i is the VWA efficiency, b_i is the branching ratio and $CF_i$the line correction factor. Since the objective is to force the U-235 activity the same for each line we have the differential attenuation relationship between gamma energy line *i* and line *j*,

$$\frac{P_i \cdot \epsilon_j \cdot b_j}{P_j \cdot \epsilon_i \cdot b_i} = \frac{CF_j}{CF_i} \qquad \text{Eq. 8}$$

The correction factors are then functions of the mass attenuation coefficients, μ, density, ρ, and average thickness, t, of the material,

$$CF_i = CF(\mu_i, \rho, t) \qquad \text{Eq. 9}$$

Everything is measured or known except ρ or t and it makes no sense to determine individually so the product $\rho \cdot t$ is defined as the unknown,

$$CF_i = CF(\mu_i, \rho \cdot t) \qquad \text{Eq. 10}$$

Suitable models for $CF(\mu_i, \rho \cdot t)$ can be found [7] for spherical, box and cylindrical shapes but a simple model approximating the spherical model can be assumed. Assuming the transmission T were known, then the correction factor for objects in where the transmission is known [6] is,

$$CF(\kappa, T) = \frac{-\kappa \cdot lnT}{1 - T^{\kappa}} \qquad \text{Eq. 11}$$

It is customary to measure T but for emission only assays T is taken as,

$$T = e^{-\mu \cdot \rho \cdot t} \quad \text{Eq. 12}$$

Which leads to, by substitution,

$$\frac{P_i \cdot \epsilon_j \cdot b_j}{P_j \cdot \epsilon_i \cdot b_i} = \frac{\mu_j \cdot (1 - e^{-\kappa \cdot \mu_i \cdot \rho \cdot t})}{\mu_i \cdot (1 - e^{-\kappa \cdot \mu_j \cdot \rho \cdot t})} \quad \text{Eq. 13}$$

The method involves taking all possible peak pair ratios for U-235 and U-238, R_{ij}, where,

$$R_{ij} = \frac{P_i \cdot \epsilon_j \cdot b_j}{P_j \cdot \epsilon_i \cdot b_i} \quad \text{Eq. 13}$$

and solve the weighted, non-linear least squares, set of *M* equations,

$$\chi^2(\rho \cdot t) = \sum_{i=1}^{M} \sum_{j=i+1}^{M} \frac{\left(R_{ij} - \frac{\mu_j \cdot (1 - e^{-\kappa \cdot \mu_i \cdot \rho \cdot t})}{\mu_i \cdot (1 - e^{-\kappa \cdot \mu_j \cdot \rho \cdot t})} \right)^2}{\sigma_{R_{ij}}^2} \quad \text{Eq. 14}$$

where,

$$\sigma_{R_{ij}}^2 = R_{ij}^2 \cdot \left(\frac{\sigma_{p_i}^2}{P_i^2} + \frac{\sigma_{p_j}^2}{P_j^2} \right) \quad \text{Eq. 15}$$

The equation set is then solved by minimizing $\chi^2(\rho \cdot t)$ with respect to the product, $\rho \cdot t$. The engine (method) has the following steps,

- Loop over the peak search records identifying peaks slated for weighted mean activity, forming the R_{ij} separately for both U-235 and U-238.
- Merge the R_{ij} for both U-235 and U-238
- Numerically solve the minimization for $\chi^2(\rho \cdot t)$ for the merged R_{ij} as well as the separate U-235 and U-238 R_{ij}.
- Compute the variance in $\rho \cdot t$, $\sigma_{(\rho \cdot t)}^2$, using standard numerical methods [8].
- Compute the activity for each line, U-235 and U-238,

$$A_{23x,i} = \frac{P_i}{\epsilon_i \cdot b_i} \cdot \frac{\kappa \cdot \mu_i \cdot (\rho \cdot t)}{(1 - e^{-\kappa \cdot \mu_i \cdot (\rho \cdot t)})} \quad \text{Eq. 16}$$

- Compute the variance for each line,

$$\sigma_{A_{23x,i}}^2 = \left(\frac{\partial A_{23x,i}}{\partial P_i} \right)^2 \cdot \sigma_{P_i}^2 + \left(\frac{\partial A_{23x,i}}{\partial (\rho \cdot t)} \right)^2 \cdot \sigma_{(\rho \cdot t)}^2 \quad \text{Eq. 17}$$

- Compute the weighted mean activity for each nuclide, U-235 and U-238.

Note there are three variants of the differential attenuation engine; **Normal**, **Apply** and **Merge**. The **Normal** method computes $\rho \cdot t$ independently for U-235 and U-238. The Apply variant allows for $\rho \cdot t$ to be computed for U-238 and applied to U-235 or *vice versa*. The **Merge** method creates a single R_{ij} for both U-235 and U-238 and computes a single $\rho \cdot t$. None of the methods require isotopics or enrichment.

Results

Multiple true waste drums, of unknown mass loadings, were measured representing a variety of waste streams and mass loadings for both rotating and multiple (static) angular and lab jack positions. More recently, the Paducah site has generated Working Reference Materials (WRM's) involving UO_2F_2 and UF_6. Currently, only rotating measurements were taken with the WRM's so only SAC (Self-Absorption Correction) and differential attenuation results are computed.

Two types of UO_2F_2 WRM's were involved in the measurements; 1"diameter x 9" height aluminum tubes packed to a density of 2.7 g/cc and 9"x7"x1/4" silicon sheet, where the UO_2F_2 powder is mixed with a silicon epoxy, called a tacky mat, which look much like a mouse pad, which is essentially diffuse. Each 1"x9" aluminum tube is packed with nominally 9 g U-235 and the tacky mats (mouse pads) contain mass loadings of about 2/3 of a gram of U-235 each. The UF_6 WRM's, bulk density of~ 5 g/cc, are called PSI tubes and consist of 1/3" copper tube bent into a U-shape (3" radius) with two copper valves attached. The nominal mass loading is for the PSI tube ranges from 1-2 g U-235 and can be 1% as well 5% enrichment.

Replicates were measured for the mock matrix sample sets shown in Table II. Three pails were configured; two alumina pellet (1.0 g/cc) and a single NaF pellet (1.24 g/cc). The sample sets were configured with the WRM's in a VWA (Volume Weighted Average) positioning so as to minimize matrix localization effects and focus on the self-absorption effects.

The replicate results, using the **Normal** mode, for both SAC and differential attenuation for each sample-matrix set are shown in Table III, Table IV and Table V for the PSI tube, mouse pad and 9" tube matrix-sample sets respectively. The assay time was 3600 s with minimal MCA dead-time. The tabulated results indicate that the differential attenuation method has quite good recovery (%R) and acceptable precision (%RSD). This is exceptional considering the 9" tube computed attenuation correction factors, shown in Table VII, are larger than a factor of 10 for the lowest energy U-235 gamma line as well as a factor of 1.3 for the U-238 1001 keV line. The mouse pad shape WRM's, as expected, exhibit the smallest correction factors whilst the UF_6 PSI tubes reveal moderate CF's about half that of the 9" tubes. This is expected as the UF_6 WRM, although denser, is quite smaller in diameter (1/3 ") versus the 9" tube (1").

Additionally, an Alumina (Al_2O_3) trap mix (true) waste drum was analyzed from the plethora of measured waste containers. The results for SAC and differential attenuation are shown in Table VI. The SAC result is 25% higher than that of the differential attenuation. A preliminary independent analysis using neutron analysis indicates that differential attenuation result is much closer to the true value. The correction factors, shown in the last column of Table VII, indicate that the Alumina trap mix waste drum self-absorption effect lies somewhere between the rather diffuse mouse pad WRM and the UF_6 PSI tube WRM.

The asymptotic nature of the infinite energy SAC model is depicted in Fig. 6 and Fig. 7 for the PPPO-WRM-C (1"x9") UO2F2 standards and the Alumina trap mix waste drum respectively.

Table II. UF_6 and UO2F2 WRM's used in the study.

PSI UF_6 Tubes			Mouse Pad UO_2F_2			PPPO-WRM-C 1"x9" UO_2F_2 Tubes		
ID	U-235 (g)	U-238 (g)	ID	U-235 (g)	U-238 (g)	ID	U-235 (g)	U-238 (g)
416	1.43	27.7	MP-4.4-001	0.645	14.1	763	9.15	174
385	1.03	20	MP-4.4-002	0.636	13.9	764	9.07	173
423	1.37	26.6	MP-4.4-003	0.623	13.6	765	9.03	172
342	1.4	27.1	MP-4.4-004	0.661	14.4	766	8.82	168
			MP-4.4-005	0.664	14.5	767	8.96	171
Total	5.23	101.4		3.229	70.5		45.03	858

Table III. PSI UF_6 tube WRM replicate (5) SAC and differential attenuation results in the alumina matrix.

	SAC	Diff. Atten.	
Replicate	U-235 (g)	U-235 (g)	U-238 (g)
1	5.77E+00	4.92E+00	9.20E+01
2	6.31E+00	4.72E+00	9.46E+01
3	6.24E+00	4.95E+00	9.68E+01
4	6.14E+00	4.78E+00	9.44E+01
5	6.30E+00	4.44E+00	9.53E+01
MEAN	6.15E+00	4.76E+00	9.46E+01
STDEV	0.226694	0.202326	1.713496
%RSD	3.7%	4.3%	1.8%
RECOVERY	117.6%	91.0%	93.3%

Table IV. Mouse pad shaped UO_2F_2 WRM replicate (6) SAC and differential attenuation results (6) in the NaF matrix.

SAC	Diff. Atten.

Replicate	U-235 (g)	U-235 (g)	U-238 (g)
1	3.32E+00	3.13E+00	6.59E+01
2	3.41E+00	3.39E+00	6.82E+01
3	3.38E+00	3.10E+00	6.89E+01
4	3.35E+00	3.13E+00	6.75E+01
5	3.29E+00	3.49E+00	6.79E+01
6	3.33E+00	3.09E+00	6.71E+01
MEAN	3.35E+00	3.22E+00	6.76E+01
STDEV	0.044862	0.172791	1.028429
%RSD	1.3%	5.4%	1.5%
RECOVERY	103.6%	99.8%	95.9%

Table V. 9" tube UO_2F_2 WRM replicate (6) and SAC differential attenuation results (6) in the alumina matrix

	SAC	Diff. Atten.	
Replicate	U-235 (g)	U-235 (g)	U-238 (g)
1	5.32E+01	4.19E+01	8.13E+02
2	5.43E+01	4.43E+01	8.22E+02
3	5.24E+01	5.09E+01	8.53E+02
4	5.53E+01	4.78E+01	8.39E+02
5	5.29E+01	4.33E+01	8.19E+02
6	5.28E+01	4.65E+01	8.37E+02
MEAN	5.35E+01	4.58E+01	8.31E+02
STDEV	1.112613	3.264333	14.98239
%RSD	2.1%	7.1%	1.8%
RECOVERY	118.8%	101.7%	96.8%

Table VI. Alumina (Al_2O_3) trap mix waste drum.

	SAC	Diff. Atten.	
Replicate	U-235 (g)	U-235 (g)	U-238 (g)
1	6.43E+01	5.09E+01	1.14E+03
2	6.47E+01	5.11E+01	1.16E+03
3	6.53E+01	5.15E+01	1.14E+03
4	6.57E+01	5.16E+01	1.15E+03
5	6.48E+01	5.19E+01	1.14E+03
6	6.30E+01	5.11E+01	1.13E+03
MEAN	6.46E+01	5.13E+01	1.14E+03
STDEV	0.916028	0.382448	10.47696
%RSD	1.4%	0.7%	0.9%

Table VII. Energy dependent Correction Factors (CF's) for the 9" tubes, PSI tubes, mouse pad shaped WRM's and Alumina trap waste drum computed from the differential attenuation method.

Nuclide	Line (keV)	UO_2F_2 9" Tubes	UF_6 PSI tubes	UO_2F_2 Mouse Pads	UO_2F_2 Al_2O_3 Trap
U-235	143.8	11.35	5.21	1.18	2.12
	163.4	8.29	3.88	1.13	1.78
	182.6	6.35	3.08	1.10	1.58
	185.7	6.10	2.98	1.10	1.55
	194.9	5.45	2.72	1.09	1.49
	202.1	5.03	2.56	1.08	1.44
	205.3	4.85	2.49	1.08	1.43
	221.4	4.13	2.21	1.06	1.36
U-238	258.2	3.06	1.82	1.05	1.25
	766.4	1.27	1.13	1.01	1.04
	786.3	1.26	1.12	1.01	1.04
	1001.0	1.20	1.10	1.01	1.03

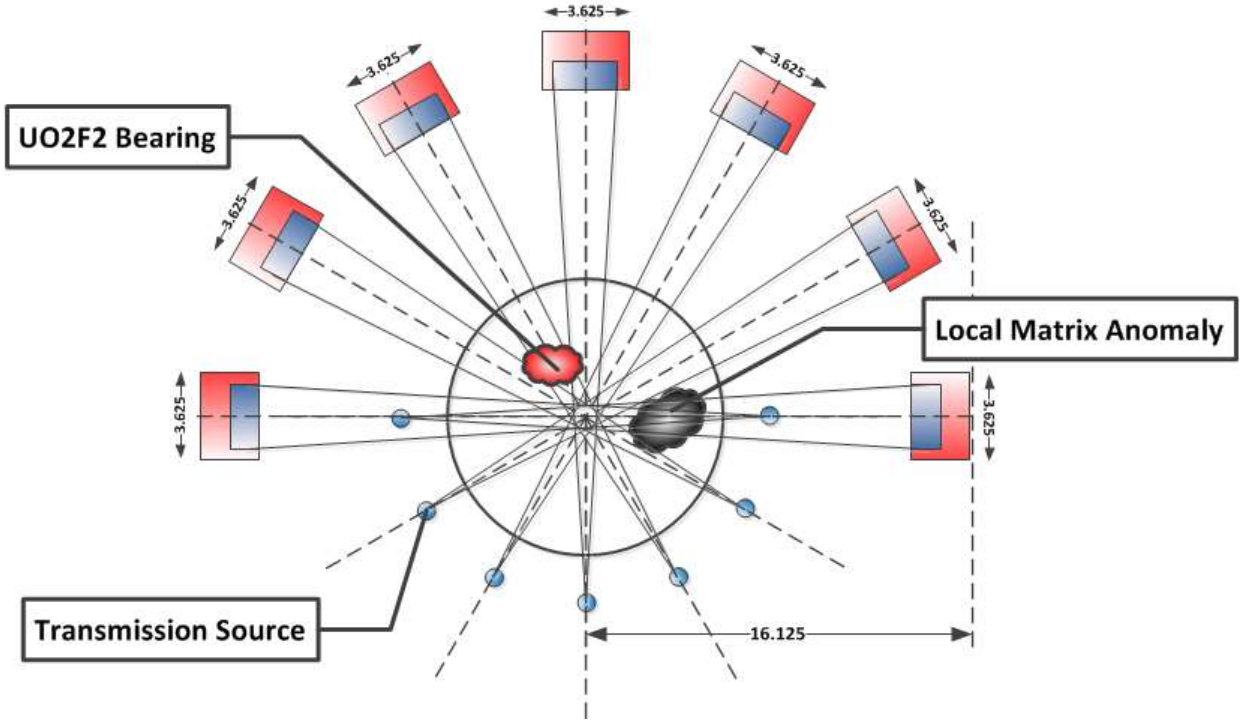

Fig. 5. Geometrical layout for acquiring data for the AIUE method. The container is rotated 0, 30, 60, 90, 120, 150 and 180 degrees with the transmission source fixed and opposing the middle detector.

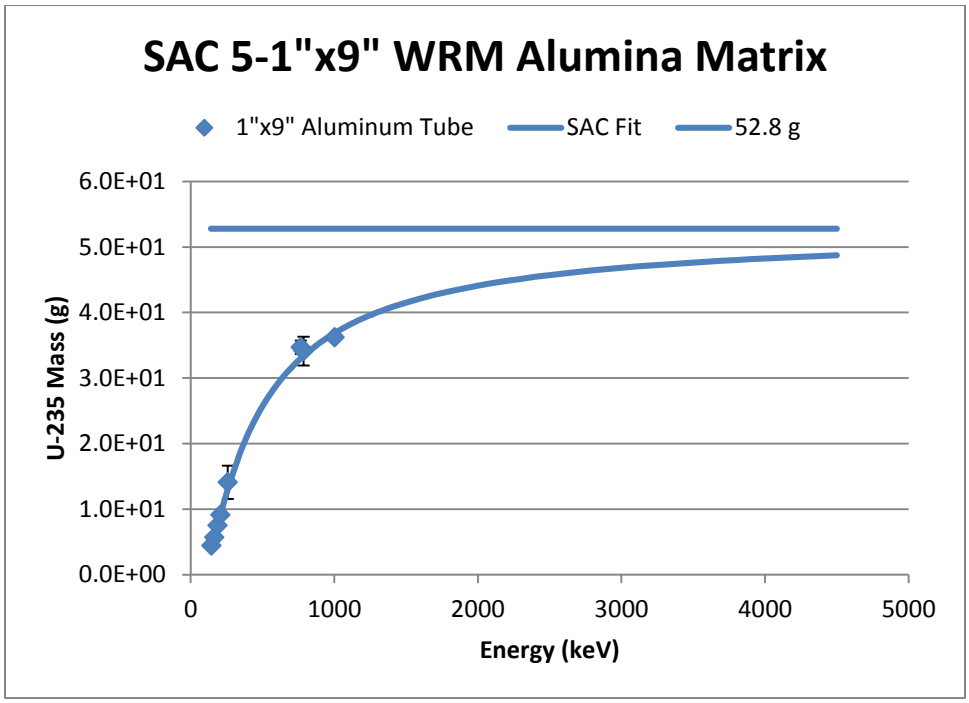

Fig. 6. The Self-Absorption Correction (SAC) curve for the PPPO-WRM-C standards (5) distributed in a Volume Weighted Average (VWA) distribution.

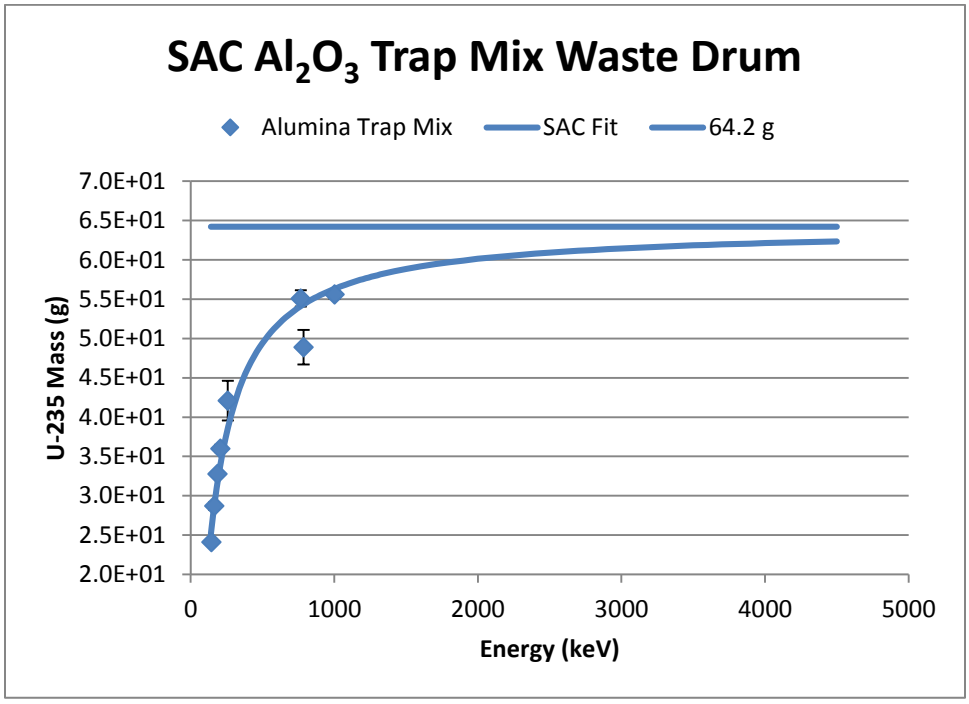

Fig. 7. The Self-Absorption Correction (SAC) curve for the Alumina (Al_2O_3) trap mix (true) waste drum.

DISCUSSION

The differential attenuation method extrudes reasonable results as long as the counting statistics are reasonable; within 10% for each line. For U-235, if the peak area of a given line happens to be greater than a two-sigma outlier then a significant bias can occur. This line can be identified by viewing the corrected mass plot and removing from the weighted mean computation in the nuclide library editor.

The three variants of the differential attenuation method; **Normal**, **Apply** and **Merge** basically produced similar results for the mock matrix drums and the Alumina trap mix waste drum, but this is mostly due to the strong counting statistics. The differential attenuation method variant utilized depends on the quality of the measured data, VWA calibration and the presence of the U-238 258.2 keV line which can be crucial. A future study will investigate the dependence of the 258.2 keV line with respect to the three variants of the differential attenuation engine. When computing $\rho \cdot t$ from the merger of U-235 and U-238 care must be taken in that the U-238 is not biased. This can be accomplished by viewing a plot of R_{ij} versus μ_i/μ_j for the merged data set.

There appears to be a positive bias associated with the Self-Absorption Correction (SAC) when compared to the differential attenuation method and the true value of the mock waste drum mass loadings. The bias can be as high as 15-25% and is most likely due to the simple model and asymptotic extraction as is evident in Fig.

6 and Fig. 7. The SAC is suitable if the measured isotopics or enrichment is stable, or well known, and serves to provide a conservative estimate if weak counting statistics are engaged. In contrast, the differential attenuation method does not utilize the enrichment but is sensitive to weak counting statistics.

The recorded recoveries (%R) and precision(%RSD) are well within the DOE NDA PDP (Performance Demonstration Plan) standards [9] which is 60-140% for recoveries and 16% for precision for low activity within an interfering matrix.

Much data has been acquired for the AIUE (Advanced ISOCS Uncertainty Estimator) method which is ideal in correcting for lumpy waste matrix. In the near future this data will be analyzed and compared to the SAC and differential attenuation methods. This study will be published in a future paper.

CONCLUSION

We have developed several new methods to minimize effects from non-uniformity, lumpy matrix and lumpy sources that exhibit severe self-attenuation and substantially deviate from VWA (Volume Weighted Avergage) calibration processes. These novel methods serve to lower the overall TMU (Total Measurement Uncertainty) budget allowing waste drums to reach Data Quality Objectives (DQO) previously unachievable. These methods have been applied to the NDA Q2 units at the Paducah Gaseous Diffusion site where data has been acquired for all the methods and analyzed for most. Preliminary results indicate that the self-absorption effects are mitigated by the SAC (Self-Absorption Correction) and differential attenuation methods where huge biases are eliminated and extremely reasonable recoveries (%R) and precisions (%RSD) were observed that would meet and exceed current DOE NDA PDP [9] and Waste Acceptance Criteria (WAC) [10] leading to proper disposition.

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