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In Situ Measurement in Low Enrichment Uranium Holdup Process Gas Piping at K-25 – Paper for Waste Management Symposia 2010 East Tennessee Technology Park Oak Ridge, Tennessee

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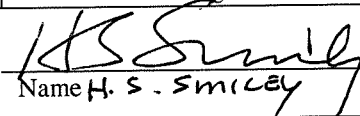
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APPROVALS

In Situ Measurement of Low Enrichment Uranium Holdup Process Gas Piping at K-25 – Draft Paper for Waste Management Symposia 2010 East Tennessee Technology Park Oak Ridge, Tennessee	BJC/OR-3357/R1
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USQD Review Determination	☒ USQD ☐ UCD ☐ CAT X ☐ Exempt (Select Criteria below.) USQD/UCD/CAT X No.: USQD – MS-REPORTS-0820, R16
Exemption Criteria	☐ (1) Non-Intent Change ☐ (2) DOE-Approved Document ☐ (3) Clearly no impact on Nuclear Facilities ☐ (4) Chief Financial Officer, Internal Audit, Labor Relations, Legal, Public Affairs, or Project Controls Organization Document
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Exhibit L Mandatory Contractor Document	☒ No (No PCCB Reviewer Signature Required) ☐ Yes (Requires review by the Proforma Change Control Board.)
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REVISION LOG		
Revision Number	Description of Changes	Pages Affected
1	Final version of paper after incorporating reviewer comments and additional refinements.	All
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FIGURES

TABLES

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ACRONYMS

BJC	Bechtel Jacobs Company LLC
DOE	U.S. Department of Energy
DQO	Data Quality Objective
HMS4	Holdup Measurement System 4
LANL	Los Alamos National Laboratory
MCA	Multi-Channel Analyzer

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EXECUTIVE SUMMARY

This document is the final version of a paper submitted to the Waste Management Symposia, Phoenix, 2010, abstract BJC/OR-3280. The primary document from which this paper was condensed is *In-Situ Measurement of Low Enrichment Uranium Holdup in Process Gas Piping at K-25 Using NaI/HMS4 Gamma Detection Systems,* BJC/OR-3355. This work explores the sufficiency and limitations of the Holdup Measurement System 4 (HMS4) software algorithms applied to measurements of low enriched uranium holdup in gaseous diffusion process gas piping. HMS4 has been used extensively during the decommissioning and demolition project of the K-25 building for U-235 holdup quantification. The HMS4 software is an integral part of one of the primary nondestructive assay (NDA) systems which was successfully tested and qualified for holdup deposit quantification in the process gas piping of the K-25 building. The initial qualification focused on the measurement of highly enriched UO_2F_2 deposits. The purpose of this work was to determine if that qualification could be extended to include the quantification of holdup in UO_2F_2 deposits of lower enrichment.

Sample field data are presented to provide evidence in support of the theoretical foundation. The HMS4 algorithms were investigated in detail and found to sufficiently compensate for UO_2F_2 source self-attenuation effects, over the range of expected enrichment (4-40%), in the North and East Wings of the K-25 building. The limitations of the HMS4 algorithms were explored for a described set of conditions with respect to area source measurements of low enriched UO_2F_2 deposits when used in conjunction with a 1 inch by ½ inch sodium iodide (NaI) scintillation detector. The theoretical limitations of HMS4, based on the expected conditions in the process gas system of the K-25 building, are related back to the required data quality objectives (DQO) for the NDA measurement system established for the K-25 demolition project.

The combined review of the HMS software algorithms and supporting field measurements lead to the conclusion that the majority of process gas pipe measurements are adequately corrected for source self-attenuation using HMS4. While there will be instances where the UO_2F_2 holdup mass presents an infinitely thick deposit to the NaI-HMS4 system these situations are expected to be infrequent. This work confirms that the HMS4 system can quantify UO_2F_2 holdup, in its current configuration (deposition, enrichment, and geometry), below the DQO levels for the K-25 building decommissioning and demolition project. For an area measurement of process gas pipe in the K-25 building, if an infinitely thick UO_2F_2 deposit is identified in the range of enrichment of ~4-40%, the holdup quantity exceeds the corresponding DQO established for the K-25 building demolition project.

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1. INTRODUCTION

The K-25 gaseous diffusion plant performed uranium enrichment from 1945 until its operational shutdown in 1985. The Bechtel Jacobs Company, LLC is in the process of decommissioning and demolishing K-25 under the U.S. Department of Energy's Accelerated Cleanup Project. In order to prepare K-25 for demolition, all of the process gas piping must be characterized, either by qualifying historical measurements where available, or by acquiring new measurements. More than 120,000 in situ measurements were made in the West Wing of K-25 to characterize the process gas piping, prior to the start of the West Wing demolition. A similar number of measurements are required for the East Wing of K-25, which is the subject of this work.

HMS4 [Smith, et. al. 2004], released in 2004, is a software package for performing and documenting holdup measurements. HMS4 can be used to measure holdup in any geometry that can be approximated by a point, line or area source. This functionality is based on the Generalized Geometry Holdup (GGH) calibration and analysis methodology developed at Los Alamos National Laboratory (LANL). HMS4 was developed as a joint effort between LANL, Oak Ridge National Laboratory and the Y-12 National Security Complex. HMS4 contains an algorithm for finite source correction to compensate for the negative bias introduced when the measured line or point source has finite dimensions which are non-negligible with respect to the field of view of the detector. In addition, there is a correction for source self-attenuation of the emitted gamma rays. HMS4 can be used in conjunction with a number of different types of scintillation detectors and multi-channel analyzers (MCA). The field measurements being performed at K-25 utilize a 1 inch diameter, ½ inch thick thallium activated sodium iodide crystal NaI(Tl) with a photomultiplier tube connected to a GBS Elektronik MCA 166.

The East Wing of K-25 was the lower enriched side of the cascade where the self attenuation correction algorithms of HMS4 can have a significant impact on the results for moderate quantities of U-235. This is driven by the increased volume of holdup material at lower enrichment, given a fixed quantity of U-235. To ensure that the continued use of the defined HMS4 detector/MCA combination would perform as expected where the average enrichment is as low as 9%, a combined theoretical and field measurement study was performed. This work also investigated the theoretical upper limit of the HMS4 holdup estimation to determine the point at which a maximum thickness of material is reached with respect to the measurement of the 185.72-keV gamma ray from U-235. It was important for the characterization process that this value fall above the nuclear criticality safety removal criteria (one of the primary factors when establishing the DQOs) for different process gas pipe sizes and components, such that the vast majority of measurements could be made without reaching maximum thickness conditions. Where maximum thickness conditions were identified it was found that the holdup estimate far exceeded the removal criteria. These cases were then handled separately using other instrumentation.

The limiting conditions for the NaI HMS4 system are characterized by a holdup mass that cannot be quantified due to the deposit presenting a mass that exceeds the maximum mass of material that the HMS4 system can identify. If this maximum mass has a corresponding quantity of U-235 which is below the DQO limit, then it is not possible to categorize the item measured as being below or above the limit and an alternate means of measurement would be necessary. Since >90% of the measurements have been, and are expected to be, below the DQO levels it was extremely important to understand the software performance and limitations in this region when characterizing uranium holdup with an enrichment range of 4-40%. The primary concern in this evaluation was to determine that under the expected in situ measurement situations the HMS4 algorithms could provide sufficiently accurate holdup measurements below the applicable decision threshold.

The average enrichment over the life of the K-25 cascade in the North and East Wings of K-25 ranges from 9.38-40.50% (excluding K-25-311-01). The established practice for HMS4 system NDA measurements in the K-25 building is to use 50% of the average enrichment over the life of the cascade within a given unit; thus, the minimum enrichment value introduced to the HMS4 algorithms will be 4.69%.

2. METHODOLOGY

HMS4 performs calculations using the generalized geometry holdup (GGH) algorithms [Russo, et. al. 2005 and Sprinkle, et. al. 1996] which allow for a measurement to be modeled as a point, line or area source. The data reduction algorithms presented here are specific to area measurements of large diameter piping (>3") which constitute the majority of the planned HMS4 measurements in the East Wing of K-25. Furthermore, it is assumed that the width of the region of interest (ROI) for the analysis peak (ROI4) and the Compton region (ROI5) are the same. For more detailed explanations of HMS4 consult the references [Russo, et. al. 2000 and Russo 2005]. Note that there may be minor notational differences between the equations presented herein and those in the references.

ROI4 defines the analysis peak region around 186-keV where the 185.72-keV U-235 gammas are expected to contribute. ROI5 is a region just above ROI4 which is used to estimate the Compton scattering contribution in ROI4. The count rate in the analysis peak (C_I) for an item measurement is given by:

$$C_I = \frac{X_{I,4} - X_{I,5}}{t_I} \text{ (counts/sec)}$$

(Equation 1)

$X_{I,4}$ = the number of counts collected in ROI4 during the item measurement

$X_{I,5}$ = the number of counts collected in ROI5 during the item measurement

t_I = the counting time for the item measurement (sec)

Similarly the count rate in the analysis peak (186-keV) for a background measurement is given by:

$$C_B = \frac{X_{B,4} - X_{B,5}}{t_B} \text{ (counts/sec)}$$

(Equation 2)

$X_{B,4}$ = the number of counts collected in ROI4 during the background measurement

$X_{B,5}$ = the number of counts collected in ROI5 during the background measurement

t_B = the counting time for the background measurement (sec)

In order to obtain the net count rate in the analysis peak, the background count rate for an area measurement of large diameter piping should be reduced to account for the attenuation of the background gamma rays due to the pipe itself.

The material attenuation correction is simply:

$$CF_{Wall} = e^{\mu_w \rho_w x_w}$$

(Equation 3)

μ_w = the mass attenuation for wall material (cm²/g)

ρ_w = the density of the wall material (g/cm³)

x_w = the thickness of the wall material (cm)

For an area source measurement of a pipe, the background corrected net count rate (C) due to the source material is given by:

$$C = \left(C_I - \frac{C_B}{e^{2\mu_w \rho_w x_w}} \right) \text{ (counts/sec)}$$

(Equation 4)

Note that when correcting the background count rate, attenuation is through 2 wall thicknesses, based on the assumption that the background gamma rays must traverse two pipe walls to be detected.

The measured U-235 areal density is then calculated from:

$$(\rho x)^{235}_{Measured} = C * K_A * CF_{wall} \text{ (g U-235/cm}^2)$$

(Equation 5)

K_A = the area calibration constant for the detector (g-s/cm^2)

CF_{wall} = the wall material attenuation correction factor

This can be converted into a measured areal density for total uranium:

$$(\rho x)^{U}_{Measured} = \frac{1}{\varepsilon}(\rho x)^{235}_{Measured} \text{ (g U/cm}^2)$$

(Equation 6)

ε = enrichment fraction

Correcting this value for self-attenuation results in:

$$(\rho x)^{U}_{True} = -\frac{1}{\mu_s}\ln\left(1 - \mu_s \cdot M_W \cdot (\rho x)^{U}_{Measured}\right) \cdot \frac{1}{M_W} \text{ (g U/cm}^2)$$

(Equation 7)

μ_s = the mass attenuation coefficient for 185.72-keV gammas in the source material, UO$_2$F$_2$ (cm^2/g).

μ_s = 1.158 cm^2/g for UO$_2$F$_2$ as specified in the HMS4 software

M_W is the molar mass ratio of UO$_2$F$_2$ to U

$$M_W = \frac{238 + 32 + 38}{238} \text{ as specified in the HMS4 software [Smith, et. al. 2004]}$$

The self-attenuation corrected value can also be put in terms of U-235:

$$(\rho x)^{235}_{True} = (\rho x)^{U}_{True} \cdot \varepsilon \text{ (g U-235/cm}^2)$$

(Equation 8)

The correction factor for self-attenuation is defined as the ratio of the true areal density to the measured areal density:

$$CF_{SA} = \frac{(\rho x)^{U}_{True}}{(\rho x)^{U}_{Measured}}$$

(Equation 9)

Using the self-attenuation correction factor, the true areal density of U-235 is:

$$(\rho x)^{235}_{True} = C * K_A * CF_{wall} * CF_{SA} \text{ (g U-235/cm}^2)$$

(Equation 10)

The mass of U-235 can then be obtained:

$$m^{235} = \frac{1}{2}(\rho x)^{235}_{True} \cdot A \text{ (g U-235)}$$

(Equation 11)

A = area associated with source (cm^2)

m^{235} = the mass of U-235 (g)

The ½ factor is introduced to account for the fact that gammas from both the near-side and far-side of the interior pipe wall (with respect to the detector position) are being counted and applied over the entire interior pipe surface area.

2.1 COUNTING UNCERTAINTY AND THE CONCEPT OF INFINITE THICKNESS

The relative counting uncertainty for an area measurement of a pipe is given below. For other geometries the background correction factor will need to be changed to the appropriate quantity [Russo, et. al. 2000].

$$\sigma_{C,relative} = \frac{1}{C} \cdot \sqrt{\frac{X_{I,4}}{t_I^2} + \frac{X_{I,5}}{t_I^2} + \frac{X_{B,4}}{\left(e^{2\mu_w\rho_w x_w}t_B\right)^2} + \frac{X_{B,5}}{\left(e^{2\mu_w\rho_w x_w}t_B\right)^2}}$$

(Equation 12)

Infinite thickness, or maximum detectable thickness, is a thickness of source material for which the maximum possible holdup estimation occurs. Adding additional amounts of source material which increase the thickness of the source (and the quantity of holdup material) will not effect the holdup estimation. This occurs due to the self-attenuation of the analysis peak gamma rays in the source material.

From equations 7 and 9, the following form for the self-attenuation correction factor can be obtained:

$$CF_{SA} = \frac{(\rho x)^U_{True}}{(\rho x)^U_{Measured}} = \frac{-\frac{1}{\mu_s}\ln\left(1 - \mu_s \cdot M_W \cdot (\rho x)^U_{Measured}\right) \cdot \frac{1}{M_W}}{(\rho x)^U_{Measured}}$$

(Equation 13)

When the self attenuation effect is negligible ($CF_{SA} \approx 1$), the true areal density is approximately equal to the measured areal density. There is a large set of physical configurations and holdup material properties for which the use of the self-attenuation correction is an adequate means for adjusting the measurement for this effect. However, due to the exponential function there is a limiting value for self-attenuation which corresponds to a maximum thickness of material. From Equation 13, it is evident that this infinite thickness occurs when:

$$\mu_s \cdot M_W \cdot (\rho x)^U_{Measured} = 1$$

(Equation 14)

Obviously, this is the limiting case for which the measured holdup quantity can no longer be estimated. This occurs for a measured areal density of total uranium of 0.67g/cm^2.

In practice, a maximum thickness warning should be produced when a particular measurement is nearing this limiting condition. P.A. Russo, et. al. [2000], recommend the following screening algorithm for the rejection of a measurement:

$$\mu_s \cdot M_W \left((\rho x)^U_{Measured} + k\sigma \left\{ (\rho x)^U_{Measured} \right\} \right) \geq 1 \qquad \text{(Equation 15)}$$

Where:

$$\sigma \left\{ (\rho x)^U_{Measured} \right\} = \sigma_R (m_A) \cdot (\rho x)^U_{Measured} \qquad \text{(Equation 16)}$$

$\sigma_R (m_A) = $ the relative uncertainty in the specific mass, m_A

$\sigma_R (m_A) = \sigma_{C,relative} = $ the relative error propagated from counting statistics (Equation 12)

The screening condition can be written as:

$$\mu_s \cdot M_W \cdot (\rho x)^U_{Measured} \left(1 + k\sigma_{C,relative} \right) \geq 1 \qquad \text{(Equation 17)}$$

This screening condition essentially moves the threshold from its theoretical limit by a factor of $k\sigma$ to account for uncertainty (typically, k=3 [Russo, et. al. 2000]).

2.2 LIMITING AREAL DENSITIES

Following work that is described in Russo, 2005 it is possible to define the maximum deposit thickness (areal density) using criteria derived from random uncertainty. This maximum deposit thickness (infinite thickness) is limited by the sum of a measured areal density plus random uncertainty as it approaches the limiting quantity of 1. Initially ignoring the component of uncertainty, the maximum values for the areal densities are given by:

$$(\rho x)^{U,MAX}_{Measured} = \frac{1}{\mu_s \cdot M_W} \qquad \text{and} \qquad (\rho x)^{235,MAX}_{Measured} = \frac{\varepsilon}{\mu_s \cdot M_W} \qquad \text{(Equation 18)}$$

An equation can be written to describe the maximum measureable areal density $(\rho x)^{U,MAX}_{Measured}$ as the sum of a limiting measurable areal density $(\rho x)^{U,LIM}_{Measured}$, and an uncertainty term:

$$(\rho x)^{U,MAX}_{Measured} = (\rho x)^{U,LIM}_{Measured} + k \left(\sigma_R (m_A) \cdot (\rho x)^{U,LIM}_{Measured} \right) \qquad \text{(Equation 19)}$$

To simplify the notation: $\sigma_R (m_A) = \sigma_{C,Relative} = \sigma$

$$(\rho x)^{U,MAX}_{Measured} = (\rho x)^{U,LIM}_{Measured} \cdot \left(1 + k\sigma \right) \qquad \text{(Equation 20)}$$

$$(\rho x)_{Measured}^{U,LIM} = \frac{(\rho x)_{Measured}^{U,MAX}}{(1+k\sigma)}$$

(Equation 21)

Substitution from Equation 18 provides:

$$(\rho x)_{Measured}^{U,LIM} = \frac{1}{\mu_s \cdot M_W \cdot (1+k\sigma)} \qquad \text{and} \qquad (\rho x)_{Measured}^{235,LIM} = \frac{\varepsilon}{\mu_s \cdot M_W \cdot (1+k\sigma)}$$

(Equation 22)

Converting from a measured quantity to a true quantity (following Equation 7):

$$(\rho x)_{True}^{U,LIM} = -\frac{1}{\mu_s \cdot M_W} \ln\left(1 - \mu_s \cdot M_W \cdot (\rho x)_{Measured}^{U,LIM}\right)$$

(Equation 23)

Substitution provides:

$$(\rho x)_{True}^{U,LIM} = -\frac{1}{\mu_s \cdot M_W} \ln\left(1 - \frac{1}{1+k\sigma}\right)$$

(Equation 24)

Figure 2.1 shows the relationship between the limiting areal density of total uranium and relative uncertainty. A practical value for the random relative uncertainty as reported in Russo, 2005 is 0.2, along with a value of k=3. Using these parameters, the limiting corrected areal density for total uranium is $0.655g/cm^2$. This is the density which is equivalent to the maximum thickness of material which can be measured using the HMS4 software. At this level and beyond, the HMS4 algorithms will report an infinite thickness; thus, applying the limiting corrected areal density over the holdup source area results in an *infinite thickness mass equivalent*. Note that this is based on a relative uncertainty of 0.2. In practice, the relative uncertainty is appreciably lower, especially under circumstances of higher count rates which are typical when nearing infinite thickness conditions. Based on the observed relative uncertainty values from a significant number of field measurements within the K-25 building, the analysis from this point forward assumes a relative uncertainty of 0.1. A relative uncertainty of 0.1 provides a limiting corrected areal density for total uranium of $0.978g/cm^2$.
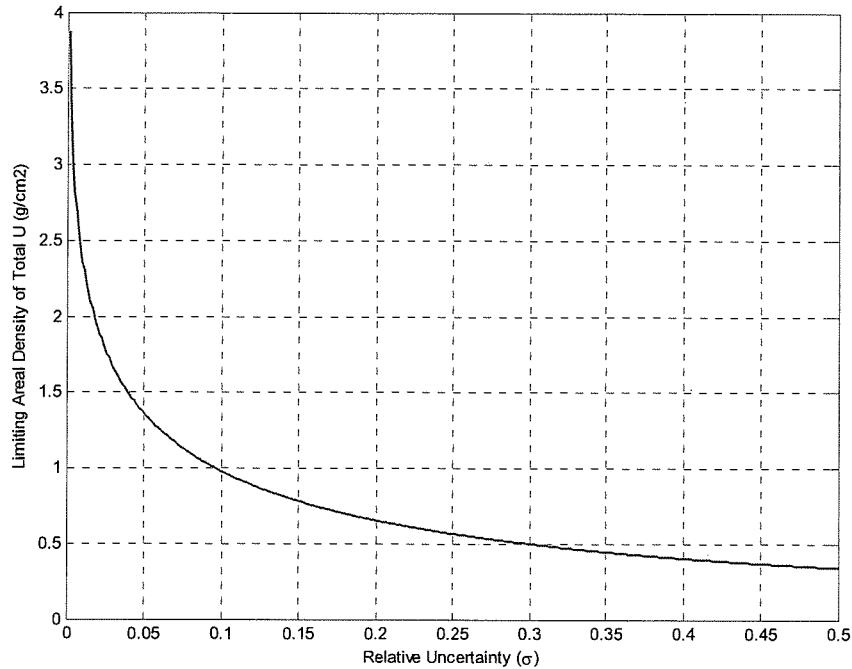
Figure 2.1. Limiting corrected areal density of total uranium (Equation 24, k=3).

Given a limiting areal density of total uranium, one can compute the limiting holdup quantities in terms of mass U-235 for a given geometry and enrichment. This quantity is referred to as the *infinite thickness mass equivalent*:

$$\textit{infinite thickness mass equivalent} \text{ (g U-235)} = (\rho x)_{True}^{U,LIM} \cdot \varepsilon \cdot A \qquad \text{(Equation 25)}$$

The most limiting expected enrichment is 4.69%. Using this enrichment, the infinite thickness mass equivalent of U-235 can be computed for the expected geometries in the East Wing of the K-25 building. Table 2.1 presents these results along with the corresponding DQOs. HMS4 should adequately quantify U-235 values below the applicable infinite thickness mass equivalent. The results in Table 2.1 indicate that when a measurement results in an infinite thickness warning from the HMS4 system, the holdup quantity sufficiently exceeds the DQO for most items with the exception of small diameter valves.

As an example, given a 10 inch diameter pipe measurement where the 50% average enrichment is 4.69%, HMS4 will adequately quantify holdup values <56.9g U-235. If an infinite thickness warning is issued for this measurement, then the expected holdup quantity is ≥56.9g U-235. Note that these conclusions along with the data in Table 2.1 are based on a relative uncertainty of 0.1. Practical results will vary with the observed relative uncertainty of the individual measurements. The infinite thickness equivalent mass will increase as the relative uncertainty of a measurement decreases. For this example, the corresponding DQO is 22 g U-235/ft (of 10 inch diameter pipe); thus, the HMS4 system can adequately characterize U-235 holdup in this geometry below the DQO mass limit.

Table 2.1. Maximum observable thick of ^{235}U based on k=3, σ=0.1, and enrichment = 4.69%

Description	Nominal Diameter (inches)	Outer Diameter (inches)	Inner Diameter (inches)	Source Surface Area A (cm²/ft)	$(\rho x)_{True}^{U,LIM}$ (g U/cm²)	ε	$(\rho x)_{True}^{235,LIM}$ (g U-235/cm²)	Infinite Thickness Mass Equivalent (g U-235/ft)	DQO (g U-235/ft)
Pipe	3	3.5	3.068	746	0.978	0.0469	0.04589	17.1	7
Pipe	4	4.5	4.026	979	0.978	0.0469	0.04589	22.5	10
Pipe	6	6.625	6.065	1475	0.978	0.0469	0.04589	33.8	14
Pipe	8	8.625	8.071	1963	0.978	0.0469	0.04589	45.0	14
Pipe	10	10.75	10.192	2479	0.978	0.0469	0.04589	56.9	22
Pipe	12	12.75	12.09	2941	0.978	0.0469	0.04589	67.5	22
Pipe	14	14	13.375	3253	0.978	0.0469	0.04589	74.6	22
Pipe	16	16	15.375	3740	0.978	0.0469	0.04589	85.8	22
Expansion Joint	3	3.5	3.37	889	0.978	0.0469	0.04589	20.4	7
Expansion Joint	4	4.5	4.37	1021	0.978	0.0469	0.04589	23.4	10
Expansion Joint	6	6.625	n/a	1301	0.978	0.0469	0.04589	29.9	14
Expansion Joint	8	8.625	n/a	1564	0.978	0.0469	0.04589	35.9	14
Expansion Joint	10	10.75	n/a	1844	0.978	0.0469	0.04589	42.3	22
Expansion Joint	12	12.75	n/a	2108	0.978	0.0469	0.04589	48.4	22
Expansion Joint	14	14	n/a	2273	0.978	0.0469	0.04589	52.2	22
Expansion Joint	16	16	n/a	2536	0.978	0.0469	0.04589	58.2	22
Valves	3	6.875	6	1876	0.978	0.0469	0.04589	43.0	40
Valves	4	6.875	6	1815	0.978	0.0469	0.04589	41.6	50
Valves	6	9.6875	8.375	3570	0.978	0.0469	0.04589	81.9	50
Valves	8	12	10.75	3567	0.978	0.0469	0.04589	81.8	50
Valves	10	13.875	12.75	5963	0.978	0.0469	0.04589	136.8	75
Valves	12	16.25	15	9957	0.978	0.0469	0.04589	228.5	75
Valves	14	17.5	16	11837	0.978	0.0469	0.04589	271.6	75
Valves	16	20.25	18.38	15921	0.978	0.0469	0.04589	365.3	75

There is one K-25 building (311-1) that has been excluded from the discussion so far due to its average expected enrichment being 0.6%, with a corresponding 50% value of 0.3%. It is not expected that HMS4 will perform well at such a low enrichment value. This does not preclude one from taking NaI measurements and analyzing the data with HMS4; however, it is expected that small amounts of uranium holdup will present infinite thickness geometry. Larger pipes with holdup quantities below the MDA of the system (typically 1-2 g U-235 for pipes between 6 and 10 inches in diameter) may be appropriately characterized but it is expected that many sections of the piping will not be quantifiable with the NaI-HMS4 system. The infinite thickness mass threshold will depend significantly on the relative uncertainty of the measurements. The conclusion that HMS4 can appropriately quantify the majority of the expected U-235 holdup in process gas piping below the DQO levels, for the K-25 East and North Wings, does not extend to building 311-1.

2.3 UNCERTAINTY IN THE SPECIFICATION OF ENRICHMENT

Rather than account for the uncertainty in enrichment specification as an individual term in the total measurement uncertainty of the HMS4 system, the enrichment values applied to field measurements are 50% of the average enrichment values over the life of the cascade. This results in a slight conservatism in the holdup estimates in most cases, except in the region where the self-attenuation correction function is highly sensitive to enrichment. In the latter cases, overestimation of holdup in the range of 30-50% (or more) can be expected.

The previous section established that U-235 areal density measurements in the region where the self-attenuation factor exhibits exponential behavior will most likely exceed the DQO. Over a significant range of enrichment, the self-attenuation correction is constant. It is only for low enrichment, or more specifically when: $\frac{\mu_s \cdot M_W}{\varepsilon} \cdot (\rho x)_{Measured}^{235} \to 1$, that the self-attenuation factor becomes extremely sensitive to enrichment.

Consider a set of simulated data as provided in Table 2.2. Based on a set of simulated analysis peak net count rates, the areal density of U-235 was calculated along with the appropriate self-attenuation correction factor. In addition, holdup mass estimates are provided for a 1 foot area measurement of 8 inch diameter pipe. Considering the effect of uncertainty in the enrichment value, it is evident from the data in Table 2.2 that over a significant range of common holdup quantities, the sensitivity of the holdup estimation to the enrichment is relatively low. It is only when the enrichment is reduced to a point which brings the self-attenuation calculation near its region of exponential behavior that significant differences in the mass estimate result from minor changes in the enrichment value. Consider that 6.52 cps in the analysis peak (8 inch pipe 1 ft section) provides 3.73 g U-235 if 4% enrichment were specified. Increasing the enrichment specification to 8% provides 3.6 g, 16% provides 3.54 g, and 32% provides 3.51 g. This example showed the minimal effect that the specification of enrichment has on the results under conditions of minor self-attenuation. Self-attenuation corrections are most significant for combinations of holdup materials and enrichments beyond what are expected, or for significant quantities of material that are well above the established DQOs.

Table 2.2. Example corrected areal densities as a function of enrichment, assuming a one foot length of 8 inch diameter pipe

C (cps)	4% U-235			8% U-235			16% U-235			32% U-235		
	CF_{SA}	$(\rho x)_{True}^{235}$ (g U-235/cm²)	m^{235} g U-235	CF_{SA}	$(\rho x)_{True}^{235}$ (g U-235/cm²)	m^{235} g U-235	CF_{SA}	$(\rho x)_{True}^{235}$ (g U-235/cm²)	m^{235} g U-235	CF_{SA}	$(\rho x)_{True}^{235}$ (g U-235/cm²)	m^{235} g U-235
0.18	1.00	5.01E-05	0.10	1.00	5.00E-05	0.10	1.00	5.00E-05	0.10	1.00	5.00E-05	0.10
1.18	1.01	3.29E-04	0.64	1.01	3.27E-04	0.64	1.00	3.26E-04	0.63	1.00	3.25E-04	0.63
1.85	1.02	5.17E-04	1.01	1.01	5.12E-04	1.00	1.00	5.10E-04	0.99	1.00	5.09E-04	0.99
2.52	1.03	7.09E-04	1.38	1.01	7.00E-04	1.36	1.01	6.95E-04	1.35	1.00	6.93E-04	1.35
2.85	1.03	8.06E-04	1.57	1.01	7.94E-04	1.54	1.01	7.88E-04	1.53	1.00	7.85E-04	1.53
3.18	1.03	9.03E-04	1.76	1.02	8.88E-04	1.73	1.01	8.81E-04	1.71	1.00	8.77E-04	1.71
6.52	1.07	1.92E-03	3.73	1.03	1.85E-03	3.60	1.02	1.82E-03	3.54	1.01	1.80E-03	3.51
13.18	1.17	4.22E-03	8.20	1.07	3.89E-03	7.56	1.04	3.75E-03	7.29	1.02	3.68E-03	7.16
23.18	1.36	8.63E-03	16.78	1.14	7.27E-03	14.13	1.06	6.78E-03	13.18	1.03	6.56E-03	12.76
33.18	1.67	1.52E-02	29.65	1.22	1.11E-02	21.65	1.10	9.98E-03	19.42	1.04	9.52E-03	18.52
66.52				1.68	3.06E-02	59.55	1.22	2.23E-02	43.42	1.10	2.00E-02	38.94
99.85							1.40	3.84E-02	74.63	1.15	3.17E-02	61.57
133.18							1.68	6.14E-02	119.36	1.22	4.47E-02	86.96
166.52							2.24	1.02E-01	199.24	1.30	5.96E-02	115.87
199.85										1.40	7.68E-02	149.43
233.18										1.52	9.74E-02	189.44
266.52										1.68	1.23E-01	238.97
299.85										1.90	1.56E-01	304.03
333.18										2.24	2.05E-01	399.02

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The over estimation in the U-235 mass due to the practice of specifying the enrichment as 50% of the average enrichment over the life of the cascade is negligible for the vast majority of HMS4 measurements since the self-attenuation correction only really starts to impact the result as the infinite thickness mass equivalent is approached. As the infinite thickness equivalent is approached the self attenuation correction begins to dominate the calculation of the *true* areal density and inaccurate enrichments can significantly impact the U-235 results. As an example, if a corrected count rate of 33.18 cps were measured and the enrichment was specified as 4% then the areal density for U-235 would be 0.0152 g/cm^2. If the true enrichment was 8%, then the areal density would have been computed as 0.0111g/cm^2; thus, the incorrect specification of enrichment resulted in an overestimation of holdup by ~37%.

The use of 50% enrichment (50% cascade life average) adequately accounts for uncertainty in enrichment specification during holdup estimations using the HMS4 system. In the majority of cases this results in a negligible overestimation; though it is also expected that in some situations the degree of overestimation may be more significant.

2.4   SIGNIFICANCE OF SELF ATTENUATION CORRECTION AT DQO LEVELS

The analysis in this section evaluates the magnitude of the self-attenuation correction as a function of enrichment for a variety of different geometries at the DQO levels established for the K-25 project. It is expected that at these DQO levels, the self-attenuation correction factor will be of reasonable magnitude (not much greater than one). Figure 2.2 provides the measured areal density as a function of the true areal density for UO$_2$F$_2$ ($\mu_s \cdot M_W$ =1.49858cm^2/g), a line with a slope of 1 is also shown for reference.
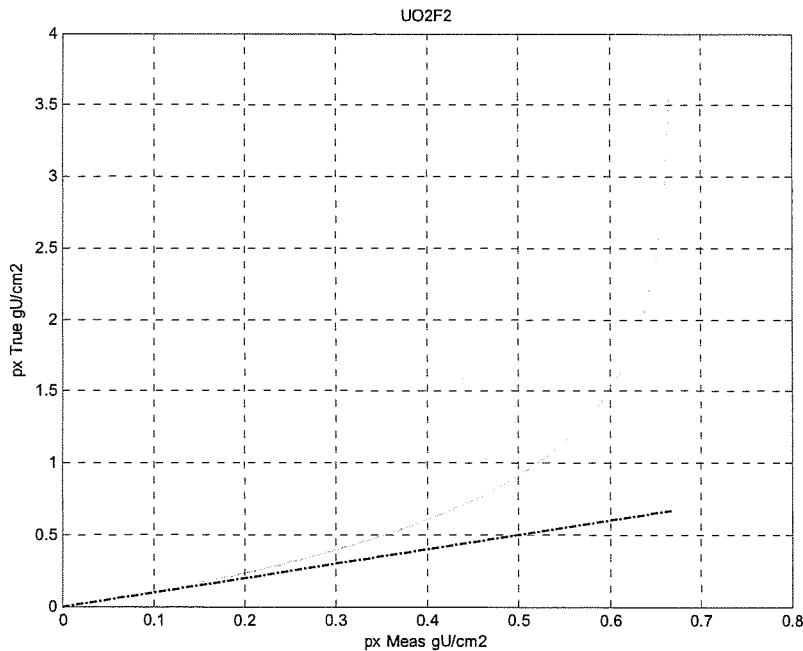

Figure 2.2. Measured U areal density as a function of True (corrected) U areal density.

Recall that the self-attenuation correction is the ratio of the true (or corrected) areal density to the measured areal density for total uranium. It is clear that below a measured areal density of total uranium of 0.2 g/cm^2 the correction is negligible; however, the HMS4 software was written to make use of the self-attenuation correction; thus, it is reasonable to expect that the HMS4 software will appropriately quantify larger density holdup quantities as well. For the purposes of the K-25 building NDA measurement campaign as long as the HMS4 system can appropriately quantify holdup quantities over a range which includes the DQO without resulting in an infinite thickness warning then the resultant measurements will be adequate for characterization tasks in the K-25 building. Items with holdup in excess of the DQOs are expected to be infrequently encountered during the measurement campaign. Figure 2.3 presents the relationship between the self attenuation correction factor and the enrichment of the UO$_2$F$_2$ deposit for a variety of pipe sizes. The relationship that is plotted in Figure 2.3 is defined by:

$$CF_{SA} = \frac{(\rho x)_{True}^{U}}{\dfrac{1}{\mu_s \cdot M_W}\left(1 - e^{-\mu_s \cdot M_W \cdot (\rho x)_{True}^{U}}\right)}$$

(Equation 26)

where $(\rho x)_{True}^{U} = \dfrac{1}{\varepsilon} \cdot \dfrac{m_{DQO}^{235}}{A_x}$

m_{DQO}^{235} is the DQO mass for the pipe of area A_x (interior surface area of a one foot section) with a fractional enrichment of ε.
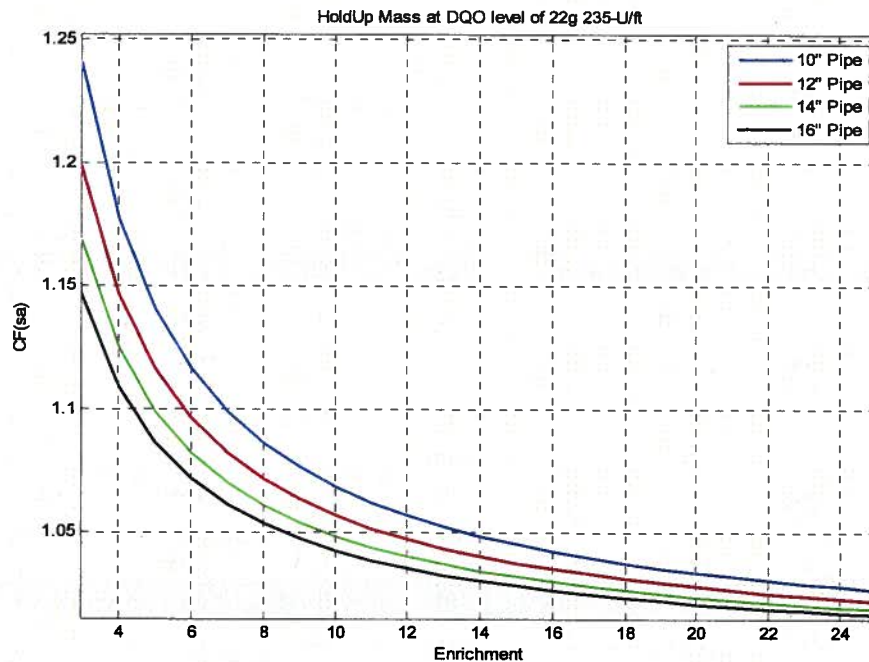

Figure 2.3. Correction factor for self-attenuation as a function of enrichment (%): 22g 235U/ft.

Down to enrichment levels of 3-4%, the DQO level (22 g U-235/ft) of material can be adequately measured and corrected for with the HMS4 algorithms, without the required self attenuation correction

factor becoming unreasonably large. These results indicate that for UO_2F_2 holdup measurements at the DQO levels established for the K-25 project and down to an enrichment of 4%, the self-attenuation correction factor remains sufficiently bound and will adequately compensate for the source self-attenuation.

2.5 FIELD MEASUREMENTS

To date >50,000 measurements HMS4 measurements have been made in the East Wing of K-25. In a few cases, HMS4 system measurements have identified infinite thickness deposits. The data for two of these cases is presented in Table 2.3. These specific sections of pipe, generically named P01 and P02, were measured specifically to evaluate the performance of the HMS4 system for a high mass, low enriched UO_2F_2 deposit. The results show that as consecutive area measurements are taken on non-overlapping one foot sections of pipe, the holdup estimate is increasing. The last quantified value for P01 is 133.23 g U-235, after which the HMS4 software reports an infinite thickness (T_∞) warning for the next four 1 foot measurements. In addition, there is a measurement labeled simulation which was artificially produced by artificially increasing the count rate to the point at which the HMS4 software would report an infinite thickness result according to equation 17.

Table 2.3. HMS4 data for K-25 building East Wing measurements

Comp. ID	NDA ID	ROI4 (cnts) $X_{I,4}$	Analysis Peak Net Rate (cps) C	Relative Uncertainty σ_R	$(px)^{235}_{Measured}$ (g U-235/cm²)	Enrichment	$(px)^{U}_{Measured}$ (g U/cm²)	$(px)^{U}_{True}$ (g U/cm²)	$(px)^{235}_{True}$ (g U-235/cm²)	U-235 (g)
P01	10	87	2.71	0.117	0.003	0.1315	0.020	0.020	0.003	3.27
	11	189	6.15	0.075	0.006	0.1315	0.045	0.046	0.006	7.55
	12	424	13.95	0.049	0.013	0.1315	0.102	0.110	0.014	17.96
	13	662	21.58	0.040	0.021	0.1315	0.157	0.179	0.024	29.21
	14	1968	64.71	0.023	0.062	0.1315	0.471	0.817	0.107	133.23
	Simulation	2621	86.48	0.020	0.083	0.1315	0.630	1.921	0.253	313.06
	15	9787	319.58	0.010	0.306	0.1315	2.327	T_∞	T_∞	T_∞
	16	8469	277.44	0.011	0.266	0.1315	2.020	T_∞	T_∞	T_∞
	17	3947	129.28	0.016	0.124	0.1315	0.941	T_∞	T_∞	T_∞
	18	4891	159.71	0.015	0.153	0.1315	1.163	T_∞	T_∞	T_∞
P02	62	430	12.94	0.056	0.012	0.075	0.159	0.182	0.014	16.88
	63	429	13.24	0.054	0.012	0.075	0.163	0.186	0.014	17.33
	64	1091	32.74	0.035	0.030	0.075	0.402	0.616	0.046	57.26
	Simulation	1614	50.18	0.028	0.046	0.075	0.616	1.716	0.129	159.53
	65	3708	107.41	0.020	0.099	0.075	1.319	T_∞	T_∞	T_∞
	66	644	18.84	0.048	0.017	0.075	0.231	0.284	0.021	26.42
	67	99	2.78	0.128	0.003	0.075	0.034	0.035	0.003	3.25

Based on the observed relative uncertainties, assumed enrichment, and source area, the infinite thickness mass equivalent of U-235 (Equation 25) can be determined for each measurement. These results are

provided in Table 2.4. For item P01, the area of each individual measurement is 2479 cm^2, the enrichment fraction is 0.1315, and k=3. Based on this, the infinite thickness mass equivalent of U-235 obtained for the simulation was 313.2g U-235 (σ = 0.02). Thus, given this level of uncertainty in the measurement the system can quantify up to 313.2g of U-235 hold up. Obviously this is well above the DQO for 10 inch diameter pipe. Similarly, the 4 measurements for P01 where the HMS4 software reported infinite thickness conditions have correspondingly high infinite thickness mass equivalents.

For item P02, the area of each individual measurement is 2479 cm^2, the enrichment fraction is 0.075, and k=3. Based on this, the infinite thickness mass equivalent of U-235 obtained for the simulation was 159.6g U-235 (σ = 0.028). Note that for both of the simulations one would expect the resultant U-235 mass estimate to agree with the corresponding maximum thickness equivalent quantity. Minor discrepancies have resulted because the calculations for each are slightly different. Most notably, the mass estimate for the simulation is based on an integer number of counts in the analysis peak region for the foreground measurement. The maximum thickness equivalent calculation is not restrained to this convention. Thus, for all practical purposes the two values (mass estimate and maximum thickness equivalent) agree.

Table 2.4. Infinite thickness mass equivalent of ^{235}U holdup for specified measurement configurations

Comp. ID	NDA ID	CF_{SA}	T_∞ Screen >1 ?	$(px)_{Measured}^{U,LIM}$ (g U/cm^2)	$(px)_{Measured}^{235,LIM}$ (g U-235/cm^2)	$(px)_{True}^{U,LIM}$ (g U/cm^2)	$(px)_{True}^{235,LIM}$ (g U-235/cm^2)	Infinite Thickness Mass Equivalent U-235 (g)
P01	10	1.015	0.040	0.494	0.266	0.898	0.118	146.378
	11	1.035	0.082	0.544	0.242	1.129	0.149	184.100
	12	1.085	0.175	0.581	0.226	1.366	0.180	222.576
	13	1.140	0.264	0.596	0.221	1.488	0.196	242.607
	14	1.734	0.755	0.624	0.211	1.829	0.240	298.075
	Simulation	3.050	1.000	0.630	0.209	1.922	0.253	313.228
	15	T_∞	3.597	0.647	0.203	2.333	0.307	380.240
	16	T_∞	3.129	0.646	0.204	2.289	0.301	373.130
	17	T_∞	1.480	0.636	0.207	2.044	0.269	333.214
	18	T_∞	1.820	0.639	0.206	2.110	0.277	343.886
P02	62	1.142	0.278	0.572	0.131	1.295	0.097	120.423
	63	1.146	0.283	0.574	0.131	1.315	0.099	122.273
	64	1.532	0.666	0.603	0.124	1.566	0.117	145.617
	Simulation	2.784	1.000	0.616	0.122	1.716	0.129	159.557
	65	T_∞	2.096	0.629	0.119	1.913	0.143	177.865
	66	1.228	0.396	0.584	0.128	1.389	0.104	129.115
	67	1.026	0.071	0.482	0.156	0.856	0.064	79.581

According to the methodology presented and quantified in Table 2.1, based on an assumed relative uncertainty of 0.1 and an enrichment fraction of 0.1315 the infinite thickness mass equivalent for a one foot section of a 10 inch diameter pipe is 159.5g U-235. Similarly, given an enrichment fraction of 0.075 and a relative uncertainty of 0.1, the infinite thickness mass equivalent for a one foot section of a 10 inch diameter pipe is 91.0g U-235. In the field examples, the observed uncertainty was lower resulting in higher infinite thickness mass equivalents than were obtained using an assumed relative uncertainty of 0.1. While this type of analysis does not provide an upper bound on the mass of material present under infinite thickness conditions it does provide a minimum expected quantity of material which in itself is useful.

3. CONCLUSIONS

A theoretical foundation was provided for the use of an HMS4-based detection system for the measurement of 185.72-keV gammas in low enriched UO_2F_2 deposits. Field data were presented, providing evidence which supported this work and its conclusions. The HMS4 algorithms were investigated in detail and found to sufficiently compensate for source self-attenuation effects over the enrichment levels of the North and East Wings of the K-25 building (~4 to 40%).

For each particular measurement there is a maximum thickness of material that HMS4 can identify, the infinite thickness mass equivalent, beyond which the HMS4 software appropriately reports an infinite thickness flag. Considering the expected enrichments of the North and East Wings of the K-25 building, the physical dimensions of the process gas pipes, and the applicable DQOs:

The process gas pipe measurement configurations will require reasonable levels of correction for source self-attenuation which HMS4 is designed to perform.

- There will be instances where the UO_2F_2 holdup mass presents an infinitely thick deposit to the NaI-HMS4 system.
- For an area measurement of process gas piping, valves or expansion joints, if an infinitely thick deposit is identified, the holdup quantity, after factoring in measurement uncertainty, will exceed the DQO.
- Collection of gamma spectra from a UO_2F_2 area source using a 1 inch diameter x ½ inch thick NaI(Tl) crystal with a photomultiplier tube connected to a GBS Elektronik MCA 166 and subsequent analysis with HMS4 is an appropriate method for the quantification of U-235 holdup in the process gas piping at K-25 over the range of applied enrichments: 4.69 – 40.50%.

This work explored the limitations and capabilities of an HMS4-based system for area measurements of process gas piping and components in the K-25 building, leading to an extension of its qualification for the quantification of U-235 holdup in UO_2F_2 deposits of lower enrichment (~4-40%). It is expected that the vast majority of the measurements in the East and North Wings of the K-25 building will be appropriately adjusted by the source self attenuation factor in HMS4. It is also anticipated that some measurements will identify infinite thickness geometry with respect to the defined HMS4 system. For these cases an alternate holdup detection system can be deployed.

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