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Modeled Uncertainty Components For The High Efficiency Neutron Counters At The Savannah River Site

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Abstract

Prior to disposal at the Waste Isolation Pilot Plant (WIPP), packages must be non-destructively assayed to verify the contents meet the WIPP acceptance criteria. As part of the acceptance criteria, twice the assay uncertainty is added to the fissile content of the package, which must be below the appropriate limit. Therefore, the assay uncertainty directly limits the quantity of fissile material that can be loaded. Experts from multiple Department of Energy laboratories have studied various measurement techniques that will give the lowest assay uncertainty on the plutonium content of Criticality Control Overpacks (CCOs) produced at the Savannah River Site.

Neutron coincidence counting was chosen as the preferred method for assay of the material. Two High-Efficiency Neutron Counters (HENCs) have been procured for these measurements. The majority of the material consists of relatively pure plutonium oxide. However, a portion of the material consists of scrap oxide containing low-Z elemental impurities, which interact with alpha particles to create additional neutrons. As a result, these impure items tend to give an erroneously high reading on the HENCs.

Monte Carlo modeling was performed to break down the total measurement uncertainty into its individual components, such as source distribution and location, chemical form, and multiplication. In particular, (alpha, n) neutrons induce additional fissions in the plutonium, resulting in a positive bias and large random uncertainty. Because the energy of these neutrons does not match the fission spectrum, these biases persist even when using multiplicity or other analysis techniques.

Plutonium-bearing materials have been sorted into families based on their elemental impurities. Working standards will be made for each family in order to determine the precision and accuracy of the HENC assay. The combination of families and working standards has been chosen to minimize the HENC uncertainty, and thus maximize plutonium loading of the CCOs.

1.0 Introduction

The Savannah River Site (SRS) will be sending plutonium-bearing material to the Waste Isolation Pilot Plant (WIPP) for disposal in a geologic repository. One of the critical criteria for acceptance of the material at WIPP is placed on the amount of Fissile Gram Equivalence mass of ^{239}Pu (FGE) within the transported Criticality Control Overpack (CCO) containers. (*Transuranic Waste Acceptance Criteria for the Waste Isolation Pilot Plant, Revision 10.0* 2020) Specifically, the measured amount of FGE plus twice its uncertainty must be less than 380 g for acceptance at WIPP. Assay measurements will be performed on each CCO using the High Efficiency Neutron Counter (HENC) to determine the ^{240}Pu-effective mass, which when combined with plutonium isotopic composition, acquired either through gamma spectroscopy of the CCO or acceptable knowledge (AK), will be used to calculate the FGE and associated uncertainty. This report documents estimates of various measurement uncertainty components relevant to determination of the FGE uncertainty.

The HENC measurement data will be evaluated with passive calibration curve analysis (CCA) to determine the ^{240}Pu-effective mass in each CCO. While it is possible to do neutron multiplicity analysis, statistical uncertainties, especially for materials with a significant amount of impurities, were estimated to be too large to satisfy the acceptance criteria for reasonable CCO mass loading. In addition to being sensitive to the fission neutrons generated by spontaneously fissioning isotopes such as ^{240}Pu and ^{238}Pu, the HENC is also sensitive to neutrons created by alpha-particle-generated neutrons from interaction with impurities in the material such as beryllium, oxygen, and fluorine. One measure of the level of impurities is α, the ratio of the neutrons generated from (α, n) reactions to the neutrons from spontaneous fission. As an example, plutonium-oxide has a α of about 1. When there are significant impurities in the material, a large number of (α, n) neutrons may be produced, which can create biases in the analysis methods.

Since there is a wide range of impurities in the 6 metric tons of plutonium to be shipped to WIPP, the material has been divided into families, pure material, impure material, very impure material and mixed uranium/plutonium, which are further divided into sub-families based either on ^{240}Pu/^{239}Pu ratio for pure materials or on the nature of the impurities for the other families, as seen in Table 1 (SRNL 2019). The grouping of the families is defined by previous knowledge of the 3013 containers, either through AK or acquired through the K-Area Measurement System multiplicity counter. Given the potential bias in the neutron measurements due to the impurities, the grouping of materials into sub-families is helpful from a perspective of understanding the potential measurement uncertainties and applying bias correction factors. The first items for measurement are to be drawn from the 1B, 2C, 2D, 3A, and 3C sub-families.

Table 1. Plutonium measurement families and sub-families

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	

The remainder of this report is divided into three additional sections. The next section presents an overview of the measurement process. The third section discusses the various contributions to the measurement uncertainty. The report ends with a conclusion.

2.0 Assay Process

The FGE in a CCO is determined from a combination of the ^{240}Pu-effective mass determined from neutron measurements and plutonium isotopic composition from either gamma spectroscopy measurements on the CCO or acceptable knowledge. In the idealized situation in which only ^{239}Pu and ^{240}Pu are present, the FGE can be calculated as

$$FGE_{idealized} = \frac{M_{240}^{eff}}{M_{240}/M_{239}}, \tag{1}$$

where M_{240}^{eff} is ^{240}Pu-effective mass determined from neutron measurements, and M_{240}/M_{239} is the ratio of ^{240}Pu to ^{239}Pu determined from either gamma measurements or AK. Uncertainties from both the neutron measurements and the plutonium isotopics will contribute to the uncertainty in FGE.

In practice, the calculation of the FGE is more complicated due to the presence of minor isotopes. The ^{240}Pu-effective mass is defined as (Ensslin et al. 1998)

$$M_{240}^{eff} = 2.54M_{238} + M_{240} + 1.68M_{242}, \tag{2}$$

where M_i are the isotopic masses for ^{238}Pu, ^{240}Pu and ^{242}Pu. The FGE, ignoring the contributions from curium and californium, is given by (US Nuclear Regulatory Commission 2013)

$$\begin{aligned} FGE = {} & 0.900M_{233} + 0.643M_{235} + 0.0150M_{237} + 0.113M_{238} + 1.000M_{239} + 0.0225M_{240} \\ & + 2.25M_{241} + 0.00750M_{242} + 0.0187M_{^{241}Am} + 34.6M_{^{242m}Am} \\ & + 0.0129M_{^{243}Am}, \end{aligned} \tag{3}$$

where the M_i values denote the isotopic mass of ^{233}U, ^{235}U, ^{237}Np, ^{238}Pu ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, ^{241}Am, ^{242m}Am, ^{243}Am, respectively as they appear in the equation. We have no process knowledge for ^{233}U, ^{237}Np, ^{242m}Am, ^{243}Am.

The first step in the evaluation of FGE is to determine the total mass of plutonium (M_{tot}). The measurement results we have at our disposal are the M_{240}^{eff} from neutron counting and plutonium isotopics, $R_i = M_i/M_{tot}$, from either AK or gamma spectroscopy. The equation for the total plutonium mass is given by

$$M_{tot} = \frac{M_{240}^{eff}}{2.54R_{238} + R_{240} + 1.68R_{242}}. \tag{4}$$

The uncertainty in the total plutonium mass is given by

$$uM_{tot} = \frac{M_{tot}}{M_{240}^{eff}} \sqrt{\left(uM_{240}^{eff}\right)^2 + M_{tot}^2\left(2.54^2 uR_{238}^2 + uR_{240}^2 + 1.68^2 uR_{242}^2\right)}, \tag{5}$$

where the prefix of "*u*" indicates uncertainties. Once the total plutonium mass is determined, the isotopic plutonium masses can be calculated from the isotopic ratios by $R_i M_{tot}$. If we define R_i for non-plutonium isotopes as also defined relative to the total mass of plutonium, we can write a compact expression for FGE as

$$FGE = M_{tot} \sum_{i=isotope} \phi_i R_i, \tag{6}$$

where ϕ_i is the isotopic weighting shown in Eqn. (3). Ignoring correlations between the total plutonium mass and the isotopic fractions, we can then write the uncertainty of FGE as

$$uFGE = \sqrt{uM_{tot}^2 + M_{tot}^2 \sum_{i=isotope} (\phi_i uR_i)^2}. \tag{7}$$

Using Eqn. (5), we can expand this equation to remove implicit terms so that

$$uFGE = M_{tot} \sqrt{\frac{\left(uM_{240}^{eff}\right)^2}{\left(M_{240}^{eff}\right)^2} + \left(\frac{M_{tot}}{M_{240}^{eff}}\right)^2 (2.54^2 uR_{238}^2 + uR_{240}^2 + 1.68^2 uR_{242}^2) + \sum_{i=isotope} (\phi_i uR_i)^2}. \tag{8}$$

The above equation shows all the terms involved in calculating the uncertainty in FGE ignoring correlations, namely the uncertainty in the ^{240}Pu-effective mass and uncertainties in the isotopic ratios.

2.1 Effective ^{240}Pu Mass

For each item, the M_{240}^{eff} is determined by neutron measurements using CCA. Two sets of standards will support this assay, Calibration Standards and Working Standards. The Calibration Standards consist of plutonium-oxide and diluent with few, if any other impurities for various amounts of M_{240}^{eff}. A set of Working Standards will be developed for each sub-family, which will consist of four or more items. All the standards are assayed with calorimetry to determine the M_{240}^{eff} with lower uncertainties than can be performed with neutron measurements. The Calibrations Standards will be used to determine the calibration of double counts to M_{240}^{eff} for items without impurities. The Working Standards will be used to estimate the range of biases in M_{240}^{eff} for each sub-family. These biases determined from the Working Standards will be used to determine a correction factor and associated uncertainty which can be applied to the CCA measurement results for each sub-family. As will be shown later, the uncertainty in the correction factor is often the leading contribution to the uncertainty in FGE for family 2 and 3 items.

2.2 Isotopics

The uncertainty due to isotopics will be drawn from AK. The measurements to support this AK are drawn from past gamma measurements as well as other assay approaches that are sensitive to isotopic mass, e.g., mass spectrometry. The uncertainties provided by the AK are assumed to be correct.

3.0 Uncertainty Estimates

To assess the uncertainty in the FGE using the HENC on each of the sub-families, several contributions must be considered. The draft TMU Summary dated Nov. 2020 divides the uncertainty contributions into two groups, one for neutron measurements to determine the ^{240}Pu effective mass and one for gamma measurements to determine isotopics. This report follows the same structure for the *neutron* measurements as the draft TMU Summary:

1. Calibration Uncertainty: passive calibration curve for the HENC using calibration standards.
2. Neutron counting statistics: random uncertainty associated with counting statistics for counting double neutron coincidences. This uncertainty will vary with sub-family.
3. Matrix/source distribution effects: physical variations in matrix mixing and fabrication of the items, positioning of the CCO, and loading of the CCO. This uncertainty will be the same for all families.
4. Background effects from local background radiation.
5. Isotopic/Chemical Form/Multiplication: item to item alpha effect variability from impurities and diluent composition differences. This uncertainty will vary with sub-family.

For the *gamma* measurement uncertainties, this report combines all the contributions listed in the TMU Summary into a single contribution described in the sixth subsection:

6. Isotopics: random variations in actual isotopic content compared to measured isotopic content.

3.1 Calibration

The calibration curve will be constructed from a set of 5 Calibration Standards. These items will be measured with calorimetry and have well-known isotopic composition such that the uncertainty on the ^{240}Pu$_{eff}$ mass will be less than 1.2%. The impact of isotopic composition uncertainty on the Calibration Standards is estimated by taking the average uncertainty in FGE due to isotopics from AK for a large subset of the items in the 6 metric tons of material. Measurements of the calibration standards will be collected with sufficient time and repetitive cycles such that the statistics uncertainty will be less than 0.5%. Under these conditions the variations between calibration standard measurements will be dominated by uncertainties associated with the variations in measurement geometry and item fabrication. Each calibration item will be composed of two cans placed into a CCO. The uncertainty resulting from placement of the CCO in the HENC is estimated to be 0.3%, the plywood composition and moisture in the CCO is expected to be at most 1.8%, and from density variations in the material is expected to be at most 1.5%. Combining these uncertainties in quadrature, results in an estimated uncertainty of 2.7% for each individual calibration standard measurement. The uncertainty resulting in a calibration curve made with the 5 standards would be estimated to be 2.5%/$\sqrt{5}$ =1.2%.

3.2 Counting

Counting statistics are expected to vary slightly with sub-families, with larger uncertainties for items with high alpha. Monte Carlo calculations estimate the statistical uncertainty for the doubles rate for an hour-long measurement to be on the order of 1 to 1.5% for families 1 and 2. For family 3 the uncertainty increase to between 2 to 4%. For each sub-family, a range of FGE mass was modeled. The precision of the measurement changes as a function of mass. The estimates quoted above are the largest uncertainties found and are considered to be an upper estimate. Table 3 shows the upper estimate count uncertainty for each sub-family.

3.3 Matrix/Source Distribution

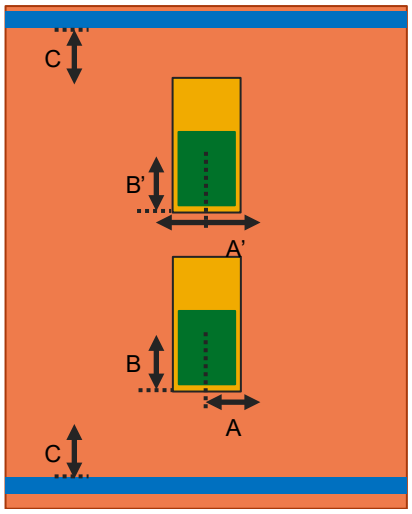

Figure 1. Conceptual diagram of CCO to explain the variation in the spatial parameters. Diagram is not to scale.

This contribution addresses how physical variations, not chemical variations, impact the measurements. Chemical variations that impact the alpha ratio are discussed in Section 3.5.2. There are a wide variety of sources that can contribute to this uncertainty. As a conservative estimate of the uncertainty contribution, the maximum bias across a range of variations modeled is reported. Information on the range of variations is listed for each term and Figure 1 will be used as a visual reference. This potential overestimate of these uncertainties may only be important for items in family 1, as the matrix/source distribution uncertainties for family 1 are comparable in magnitude to the other contributions to the uncertainties. If required, a refined analysis for family 1 can be completed later. The biases in the coincidence counts due to the described variations were investigated using an MCNP version 6.2 (*MCNP6 Version 2.0 User's Manual- Code Version 6.2* 2017) model of the HENC detector.(Siciliano, Moore, and Warren 2021) The contributions to this matrix/source distribution uncertainty are:

- Variations of the position of cans resulting from:
    - Vertical and radial position of the cans due to variations in the positioning of the bagout bag

      This uncertainty is estimated to be 0.3%. Several cases were modeled with the position of the material varied within the can/CCO. The modeling allowed the cans to move independently by 0.75″ horizontally or 0.9″ vertically, as indicated by A/A' and B/B' in Figure 1. Due to cylindrical symmetry, it is only necessary to adjust one can horizontally in one direction while the other can is adjusted to both the left and right of the axis of symmetry, see A versus A'. The worst case, which is used to determine the uncertainty for this contribution, was when the cans were moved vertically apart (component B) by a total of 1.8″.

- Variations from the plywood
    - Volume of the plywood due to fabrication tolerances
    - Moisture of the plywood due to weather/seasonal variations

      This uncertainty is estimated to be 1.8%. The plywood volume was varied from -18% to +5% and the moisture content in the plywood was varied from 0% to 20%. The volume of the plywood was adjusted by modifying its thickness, as indicated by C in Figure 1. The uncertainty for this contribution is taken from the worst-case option which is the difference between 0% and 20% moisture content.

- Inhomogeneity due to incomplete mixing

This uncertainty is estimated to be 1.9%. This uncertainty is the difference in bias between a completely homogenized mixture of plutonium and diluent and a heterogenous mixture in which 60% of the plutonium is mixed with the diluent and 40% of the plutonium is not. If no mixing occurs, a bias difference of 14% is observed.

- Diluent composition

  This uncertainty is estimated to be 1.2%. The modeling varied the diluent composition 30 times using samples from a Dirichlet distribution (10% STD on each item). No new constituents were added.

- Nickel coating thickness

  This uncertainty is estimated to be 1.0%. The nickel coating reduces the (α,n) neutrons from boron, but there are variations in the (α,n) neutron emission rates because of variations in the nickel coating thickness. The uncertainty for this component was quoted as the difference in bias between items with the nickel shielding efficiency as observed in (Aucott and Scogin 2018) compared to items in which the nickel shielding efficiency is reduced by approximately 30%. According to Sources4C, this corresponds to a change of roughly 5 µm to 1.5 µm.

- Density variation

  This uncertainty is estimated to be 1.5%. The modeling included varying the density over a range of 2.8 to 3.6 g/cm^3. The uncertainty reflects the bias difference observed between items of nominal density and 2.8 g/cm^3.

Adding these terms together in quadrature results in a 3.4% uncertainty.

The results in this report assume a loading of 50 g B_4C per CCO (25 g per can). The B_4C has two competing effects on the neutron coincidence measurements, namely that it decreases the multiplication and increases the (α,n) neutrons. Modeling results indicate that adjusting the B_4C so that there is no B_4C or 100 g B_4C in the CCO results in a bias of 3% or 1.4%, respectively, compared to items with 50 g B_4C per CCO. These impacts will be most important for family 1, which are expected to have comparable uncertainties. For the other families, the additional uncertainty introduced by this bias shift is likely to be small compared to uncertainty due to chemical composition, see Section 3.5.2. New Working Standards for family 1 items would be required if the amount of B_4C changes from the current 50 g per CCO to capture the change in the bias correction factor.

Another variation explored but not covered in detail is loading a single can in a CCO, versus the nominal case of two cans. A bias of about 4% was found if one of the cans was removed. Similar to the removal of B_4C, treating this bias as an uncertainty would have an impact mostly on CCOs for family 1. Alternatively, one could perform new calibrations or create working standards for 1 can CCOs to address this bias, if the 4% uncertainty raises significant concerns.

3.4 Background

The impact of the background on the total measurement uncertainty for the CCA method, which uses the measured doubles count rate, is expected to be negligible. Typical doubles background rates are statistically zero. Nonzero doubles background may result from coincident neutrons detected from nearby fissioning materials or neutron bursts produced from cosmic ray interactions with nearby high-Z materials. With proper background subtraction procedures, this small but non-zero background can be corrected for and the associated uncertainty will be no larger than several counts per second. The statistical uncertainty in the measured doubles from the plutonium materials will range from 1 to 4% (see section 3.2) and the doubles count rates are expected by be greater than 1000 counts per second. The resulting magnitude of the uncertainty in the measured count rates will be on the order of 10s of counts per second

which is much greater than the uncertainty resulting from the background. The background will not contribute to the total measurement uncertainty except possibly for items with very low plutonium loading, of which none are expected.

3.5 Isotopics/Chemical Form/Multiplication

This contribution addresses how the isotopic makeup and chemical form impact the neutron measurement. We assume that the isotopic impact is addressed in the gamma measurement section. The chemical form has a significant impact, as different impurities can lead to different alpha ratios, as well as different emitted neutron energy distributions, which will impact the efficiency of the detector. One thing to note is that only sub-families 1C and 1D considered non-WGPu; all other families only considered WGPu in the Monte Carlo calculations. If non-WGPu is considered for the other sub-families, the biases used to determine the uncertainties may change for those sub-families.

3.5.1 Pu Isotopics

See Section 2.2 which discusses the contributions of the plutonium isotopics to the uncertainty of the FGE.

3.5.2 Chemical Composition

This section addresses variations in the measurements due to variations in the chemical composition (amount of low-Z impurities in the item). Increasing concentrations of low-Z impurities will lead to the production of additional neutron through the (α, n) processes. The additional neutrons will induce extra fissions resulting in a mass bias. Each sub-family has a range of alpha values and low-Z elements which will bias the measurement result. This bias can be addressed through the use of the Working Standards to develop a bias correction for each sub-family. However, variations in the amount of low-Z impurities (or alpha value) within the sub-families will mean that a single bias correction for the entire sub-family will introduce some uncertainties. The following assumes a constant bias correction for each sub-family without additional α-dependent corrections. A summary of the evaluation process is provided in (PNNL-31054, Appendix A). Uncertainties predicted from the Monte Carlos calculations are summarized in the table below, which assume a uniform distribution of alpha values within the sub-family.

Table 2. Uncertainties per sub-family due to variations in chemical composition

Sub-family	α range	Uncertainty (%)
1A	1-3	1.4
1B	1-3	1.0
1C	1-3	1.9
1D	1-3	2.8
2A	1-10	6.8
2B	1-10	6.8
2C	3-10	6.8
2D	3-10	6.7
2E	3-10	6.7
3A	10-100	59.2
3B	10-80	34.2
3C	10-50	51.1
3D	10-20	12.2
3E	10-20	14.0

3.6 Isotopics

This contribution addresses how uncertainties in the relative isotopic masses may impact the uncertainties in FGE. The isotopic uncertainties will be drawn from AK, which nominally is derived from gamma measurements but may come from other assay approaches as well. We can estimate the uncertainties in the FGE from the isotopics using historical isotopic information for roughly 1400 containers that will be processed through the disposal process. We determined the average uncertainty and its standard deviation across these containers weighted appropriately by the factors in Eqn. (8). To provide a conservative estimate of the isotopic uncertainty contribution, we use the average plus the standard deviation. Of note is that Eqn. (8) is not an expression of relative uncertainty, since the factor in front of the square root is M_{tot} and not FGE. We assume $FGE/M_{tot} = 1$ to conservatively estimate the isotopic contribution. We estimated only the dominate contributions to the uncertainty in FGE from the isotopic information:

- ^{238}Pu: 0.1%
- ^{239}Pu: 0.4%
- ^{240}Pu: 1.6%
- ^{241}Pu: 0.1%
- ^{242}Pu: 1.0%

Added in quadrature, these suggest an uncertainty of 2.0% in the FGE due to uncertainties in the isotopics. This uncertainty is based on all ~1400 items, without regard to the family of the item. There may be small variations among sub-families. As this term will not be the leading term in the overall uncertainty, we have opted not to study this component of the uncertainty per sub-family.

4.0 Conclusions

Table 3 summarizes the estimated contributions to the measurement uncertainty of the FGE. Uncertainties of 1.2%, 3.4%, 0% and 2.0% were used for calibration, matrix/source distribution, background and isotopics for all sub-families.

Table 3. Uncertainty Budget for Total Measurement Uncertainty of FGE.

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

5.0 References

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