Calorimeter-Based Adjustment of Multiplicity Determined $^{240}Pu_{eff}$: Known-α Analysis for the Assay of Plutonium

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N-TRT-K-00020

Savannah River Nuclear Solutions

Revision 0
February 21, 2012

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Introduction

In nuclear material processing facilities, it is often necessary to balance the competing demands of accuracy and throughput. While passive neutron multiplicity counting is the preferred method for relatively fast assays of plutonium, the presence of low-Z impurities (fluorine, beryllium, etc.) rapidly erodes the assay precision of passive neutron counting techniques, frequently resulting in unacceptably large total measurement uncertainties. Conversely, while calorimeters are immune to these impurity effects, the long count times required for high accuracy can be a hindrance to efficiency.

The higher uncertainties in passive neutron measurements of impure material are driven by the resulting large (>> 2) α-values, defined as the (α,n):spontaneous fission neutron emission ratio. To counter impurity impacts for high-α materials, a known-α approach may be adopted. In this method, α is determined for a single item using a combination of gamma-ray and calorimetric measurements. Because calorimetry is based on heat output, rather than a statistical distribution of emitted neutrons, an α–value determined in this way is far more accurate than one determined from passive neutron counts. This fixed α value can be used in conventional multiplicity analysis for any plutonium-bearing item having the same chemical composition and isotopic distribution as the original. With the results of single calorimeter/passive neutron/gamma-ray measurement, these subsequent items can then be assayed with high precision and accuracy in a relatively short time, despite the presence of impurities.

A calorimeter-based known-α multiplicity analysis technique is especially useful when requiring rapid, high accuracy, high precision measurements of multiple plutonium bearing items having a common source. The technique has therefore found numerous applications at the Savannah River Site. In each case, a plutonium (or mixed U/Pu) bearing item is divided into multiple containers. A single item from that batch is then selected for both neutron and calorimetric measurements; all remaining items undergo a neutron measurement only. Using the technique mentioned above, the "true" α value determined from the first (calorimeter and passive neutron measured) item is used in multiplicity analysis for all other items in the batch. The justification for using this α value in subsequent calculations is the assumption that the chemical composition and isotopic distribution of all batch items are the same, giving a constant (α,n):spontaneous fission ratio.

This analysis method has been successfully applied to the KIS Facility, significantly improving measurement uncertainties and reducing processing times for numerous items. Comprehensive plans were later developed to extend the use of this method to other applications, including the K-Area Shuffler and the H-Area Pu-Blending Project [1,2,3]. While only the feasibility study for the Shuffler has been completed, implementation of the method in the H-Area Pu-Blending Project is currently in progress and has been successfully applied to multiple items. This report serves to document the details of this method in order to serve as a reference for future applications. Also contained herein are specific examples of the application of known-α multiplicity analysis.

Method Overview

As part of the nondestructive assay of an item containing plutonium or a mixture of plutonium and uranium, the item first undergoes a gamma-ray spectroscopy measurement to determine the isotopic distribution of the nuclear material [4]. Supplementing the gamma-ray measurement is a second nondestructive technique focusing on a specific material property. Analysis of the combined results then yields plutonium content. Similar results can also be determined for the uranium content, though this is a more indirect measurement depending on the uranium/plutonium ratio and declared enrichment.

Calorimetric measurements are widely accepted as the preferred complementary method for high accuracy, high precision plutonium assay [5]. However, given the long (~ 8+ hours) count times required for a typical measurement, a second option is the use of passive neutron multiplicity counting, capable of yielding plutonium mass uncertainties of < 10 % in ~ 1 hour when using a high efficiency neutron counter [6]. Though measurement time is shorter, a significant drawback to the use of passive neutron counting is that it relies upon accurate assessment of the statistical distribution of emitted neutrons. Random emissions such as those which may originate from (α,n) reactions can skew these distributions, directly affecting results. Neutron detection techniques are thus inherently sensitive to impurities, which contribute to these random events.

A third option exists which combines these three instruments [7]. This combination is typically used when neutron measurement uncertainties exceed desired thresholds, impurities or instruments inhibit a reliable neutron measurement, or extensive calorimetric measurements are not conducive to operational throughput requirements. The combined method specifically targets the (α,n):spontaneous ratio (α)calculated in passive neutron measurements. By focusing on α, the term most affected by material impurities, expected or observed biases in the plutonium (and relatedly the uranium) mass values can be offset.

In order to discuss calorimeter based known-α multiplicity analysis, it is first necessary to explain the two combined methods used to determine plutonium content. As mentioned above, quantification of plutonium content typically begins with analysis of gamma-ray spectra, collected using a co-axial high purity germanium (HPGe) detector. The spectra are analyzed to determine the distribution of all plutonium isotopes of interest, along with the other commonly occurring nuclides; concentrations of these additional nuclides (^{235}U, ^{238}U, ^{237}Np, and ^{241}Am) are normally specified with respect to total plutonium content. The second portion of the measurement may utilize either neutron multiplicity counting, used to determine the rate and temporal distribution of neutron emission, or calorimetry, which measures heat output.

Neutron Multiplicity Counting: Determination of $^{240}Pu_{eff}$

As the nuclides present in a given item undergo spontaneous and/or induced fission, the neutrons are emitted in bursts. The detected counts are binned in time to determine the uncorrelated (singles) and correlated (doubles, triples, etc.) neutron emission rates. Multiplicity analysis requires accurate measurement of these distributions, which serve as inputs to the multiplicity equations. A description of all terms in the multiplicity equations is given in Table 1[6].

Term	Definition
F	^{240}Pu spontaneous fission rate
ε	neutron detection efficiency
M	neutron leakage multiplication
α	(α,n):spontaneous fission neutron ratio
$\nu_{s1}, \nu_{s2}, \nu_{s3}$	first, second, and third spontaneous fission moments
$\nu_{i1}, \nu_{i2}, \nu_{i3}$	first, second, and third induced fission moments
f_d	doubles gate fraction
f_t	triples gate fraction

Table 1 Definition of variables/constants used in the neutron multiplicity equations

Though the analysis software/hardware common to neutron counters allows resolution of multiplicities well beyond triply correlated events, conventional multiplicity analysis uses only the singles, doubles, and triples rates. However, higher measured multiplicities are inputs to the calculated singles rate. The singles neutron rate equation,

1)
$$S = F\varepsilon M\nu_{s1}(1+\alpha)$$

represents the total contribution of all neutrons detected, i.e. neutrons originating from spontaneous and induced events. No distinction is made between spontaneous and induced fission events.

The doubles rate equation (Equation 2) contains terms related to emitted spontaneous fission neutrons, and also distinguishes between fissions induced by the emission of spontaneous neutrons, and fissions induced by the multiplication of (α,n) neutrons. The doubles equation thus accounts for both first and second order events which can lead to the detection of doubly correlated neutrons.

$$D = \frac{F\varepsilon^2 f_d M^2}{2}\left[\nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{s1}(1+\alpha)\nu_{i2}\right]$$

2)

The triple coincidence or triples rate defined in Equation 3 is similar to the doubles rate equation in that it accounts for both first and second order neutron source events. However, as there are several combinations of events which can lead to this type of correlation, the equation accounts for the many multistep processes which can lead to simultaneous neutron emission.

3)

$$T = \frac{F\varepsilon^{3} f_{t} M^{3}}{6}\left\{\nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right)\left[3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}\right] + 3\left(\frac{M-1}{\nu_{i1}-1}\right)^{2} \nu_{s1}(1+\alpha)\nu_{i2}{}^{2}\right\}$$

It is important to note that Equations 2 and 3 contain the gate fraction terms f_d and f_t respectively. These scaling factors denote the fraction of detected double or triple coincidence events. Also, because analysis of the rates and distributions must take into account the origins (both spontaneous and induced fissions) of the distributions, certain analysis parameters, namely the induced fission moments, are expressed as functions of material makeup [8].

Determining information about the plutonium content first requires solving the above multiplicity equations for M, the sample multiplication. This is done by iteratively solving

4)

$$a + bM + cM^{2} + M^{3} = 0\,,$$

where a, b, and c are functions of the various multiplicity analysis parameters. Once a value for M is determined, the fission rate of the item, given by

5)

$$F = \left(\frac{1}{\varepsilon M^{2}\nu_{s2}}\right)\cdot\left[\frac{2D}{\varepsilon f_{d}} - \frac{M(M-1)\nu_{i2}S}{\nu_{i1}-1}\right]$$

is determined.

In these items, the primary neutron source is spontaneous fission of the even nuclides present, with ^{240}Pu being the greatest contributor. The masses of all spontaneous neutron sources are therefore normalized to the ^{240}Pu mass, giving the following expression for ^{240}Pu$_{\mathrm{eff}}$,

6)

$$^{240}Pu_{eff} = 2.52m_{238} + m_{240} + 1.68m_{242}$$

where m_{238}, etc. are the masses of ^{238}Pu, ^{240}Pu, and ^{242}Pu respectively. Note that Equation 6 only lists the even isotopes of plutonium (^{238}Pu, ^{240}Pu, ^{242}Pu) as contributors to the neutron emission spectrum. While normalization factors do exist for the other nuclides present, their spontaneous fission contributions are negligible to the analysis.

Because ^{240}Pu$_{\mathrm{eff}}$ represents the sum of all neutron sources and is directly related to the neutron emission rate of ^{240}Pu, the spontaneous fission mass for a measured item is determined by dividing the measured rate F by the emission rate/gram of ^{240}Pu. Thus,

7) $$^{240}Pu_{eff} = \frac{F}{F_{240}},$$

where F_{240} = 473.5 n/s·g. The total plutonium mass is then given by,

8) $$Pu_{Total} = \frac{^{240}Pu_{eff}}{2.52 f_{238} + f_{240} + 1.68 f_{242}}$$

with the f_n's representing the isotopic percentages determined from gamma ray measurements. The mass of each nuclide is then determined from a combination of the isotopic distribution and total plutonium mass.

Calorimetric Determination of Total Plutonium

Both neutron-based and calorimetric measurements of plutonium depend upon nuclear phenomena, i.e. radioactive decay. However, rather than directly measuring the decay products (neutrons, α-/β-particles, or γ-rays), calorimeters measure the energy released from these decays and any subsequent reactions. The driving principle behind calorimeter operation is the assumption that nearly all of this decay energy is released as heat. A calorimetric measurement depends upon the transfer of this heat energy from one system, i.e. the sample of interest, to a well characterized measurement system. The method therefore quantifies the total output of an entire item. As such, calorimetric assays are independent of the geometry, material distribution, and impurity concerns that may affect a neutron based measurement.

Nuclide	Specific Power (mW/g)	Specific Power Std. Dev (mW/g)
^{238}Pu	567.570	0.260
^{239}Pu	1.929	0.003
^{240}Pu	7.082	0.002
^{241}Pu	3.412	0.002
^{242}Pu	0.116	0.003
^{241}Am	114.200	0.420

Table 2 Specific power factors for the nuclides of interest in special nuclear material [2]

The specific power factors for the typical nuclides of interest in special nuclear material (SNM) are given in Table 2. From the standard deviations shown, which are typically much less than 1 %, it is clear that the specific powers associated with each of the listed nuclides are very well quantified. This stems from the relatively short half-lives of plutonium and ^{241}Am, allowing for a high rate of power generation. The isotopes of uranium are omitted from Table 2 as these isotopes are very long lived, resulting in a negligible contribution to the total heat output. Calorimetric measurements are therefore not a viable option for direct uranium assay.

Radioactive decay of each of the listed nuclides is unaffected by the other decaying species. Therefore, the heat of each nuclide's decay contributes independently to the total heat output of a given sample. Specifically, for a sample with known isotopic distribution, an effective power can be defined such that

9)
$$P_{eff} = \sum_i p_i f_i \,,$$

where p_i is the specific power for a particular decaying nuclide and f_i is the respective mass fraction.

Given a plutonium sample with known isotopic distribution and a measured power output W, the total plutonium is given as

10)
$$M = \frac{W}{P_{eff}} \,,$$

where sample power is measured in watts. As in passive multiplicity assays, the mass of each nuclide is determined from a combination of the isotopic distribution with total plutonium mass. While it is straightforward to calculate total plutonium in this way, a single calorimeter measurement typically requires 8 – 12 hours.

The Calorimeter-Based Known-α Methodology

Determination of $^{240}\mathrm{Pu}_{\mathrm{eff}}$ using passive neutron multiplicity counting depends on the sample having relatively few impurities, as this results in less interference in distinguishing correlated and uncorrelated neutron emission events. When this impurity condition is met, the (α,n):spontaneous fission neutron ratio can be accurately quantified. Because this ratio is a crucial component in all three of the multiplicity equations, when the impurity condition is not met, the measured $^{240}\mathrm{Pu}_{\mathrm{eff}}$ mass may have large uncertainties or be considered unreliable. To offset this sensitivity, a calorimetric measurement can be used to more accurately determine α. Using this independently measured value for α, a more accurate adjusted $^{240}\mathrm{Pu}_{\mathrm{eff}}$ mass may be calculated.

In order to perform a calorimeter-based mass adjustment, a calorimetric measurement is first used in conjunction with the isotopic distribution to fix total plutonium, as well as the individual isotopic masses. With Equation 6, the measured masses of the even plutonium isotopes are then used to calculate a value for $^{240}\mathrm{Pu}_{\mathrm{eff}}$. Given the accuracy of the calorimeter, this calculated $^{240}\mathrm{Pu}_{\mathrm{eff}}$ mass is considered the "true" value, as it is immune to neutron effects. Based on the assumption of accuracy, this $^{240}\mathrm{Pu}_{\mathrm{eff}}$ is then used in Equation 7 to calculate the actual spontaneous fission rate.

Recall that Equation 5 relates the spontaneous fission rate to several variables, including the sample leakage multiplication. Using the calorimeter-determined fission rate, an actual leakage multiplication can be calculated. This calorimeter-adjusted value M_C is determined from Equation 11,

11)
$$\left[F\varepsilon^2 f_d \frac{\nu_{s2}}{2} + \frac{\varepsilon f_d \nu_{i2} S}{2(\nu_{i1}-1)}\right] M_C^{\,2} - \frac{\varepsilon f_d \nu_{i2} S}{2(\nu_{i1}-1)} M_C - D = 0\,,$$

which is simply a rearrangement of Equation 5. Though this is a quadratic equation, only one solution is accepted as a negative leakage multiplication has no physical meaning. Once M_C has been calculated, the calorimeter-adjusted value for α is determined using a rearrangement of Equation 1, giving

12)
$$\alpha_C = \frac{S}{F\varepsilon \nu_{s1} M_C} - 1\,,$$

where the subscript "*C*" indicates the "true" (α,n):spontaneous fission neutron ratio. Given the high accuracy and precision of calorimetric measurements, uncertainties in α_C and M_C are typically << 1 %.

The value of α_C will likely differ from the value of α determined from passive neutron counting. However, because the calorimetric values are free of impurity effects, this new value is used to align the calorimeter and multiplicity generated mass results.

Conventional multiplicity analysis relies on analysis of the measured neutron emission distributions to yield information about the sample leakage multiplication and subsequently the (α,n):spontaneous fission neutron ratio. However, if α is already known, the neutron emission distributions can be combined with this known value to yield a calorimeter-corrected plutonium mass. This is done by first rearranging the singles and doubles multiplicity equations (Equations 1 and 2), giving Equation 13.

13)
$$\frac{\nu_{s1}\nu_{i2}}{\nu_{s2}(\nu_{i1}-1)}(1+\alpha_C)M^2 + \left[1 - \frac{\nu_{s1}\nu_{i2}}{\nu_{s2}(\nu_{i1}-1)}(1+\alpha_C)\right]M - \frac{2D\nu_{s1}}{S\varepsilon f_d \nu_{s2}}(1+\alpha_C) = 0\,,$$

Because relative density, fill height, etc. may vary, it is still necessary to calculate a unique leakage multiplication for each item based on measured neutron emission distributions. The sample leakage multiplication determined from Equation 13 then serves as an input, along with the measured singles and doubles rate, to Equation 5, which in turn is used to calculate the spontaneous fission emission rate. Calculation of total plutonium then follows the established method for conventional multiplicity analysis. It is in this way that the accuracy and precision of a calorimeter can be achieved while only requiring a relatively short neutron measurement.

Method Uncertainty

The current error propagation scheme that was originated in the KIS Facility and extended to other applications depends primarily on the calculated calorimeter uncertainty. For the KIS calorimeter, the systematic, calibration, and random uncertainties were calculated at 0.0003, 0.0023, and 0.0073 W respectively for heat outputs of less than 1 W and 0.0020, 0.0016, and 0.0049 from 1 – 25 W [9]. An additional systematic bias of -0.78 % is also included for plutonium masses greater than 200 g, stemming from calibration of the gamma isotopic system.

For a calorimeter measurement, the -0.78 % bias is first applied to the final mass result. The error in this result takes inputs from the gamma isotopic system, written as

14)
$$\sigma_{power}^2 = \sigma_{sys}^2 + \sigma_{cal}^2 + \sigma_{ran}^2 ,$$

where the systematic, calibration, and random uncertainties were previously determined to be 0.21, 0.00, and 0.83 % respectively for plutonium masses greater than 200 g [10]. This error is propagated along with the calorimeter uncertainty giving

15)
$$\sigma_{total}^2 = \sigma_{power}^2 + \sigma_{cal}^2 ,$$

where σ_{cal} is expressed as a percentage of the measured power. This typically results in uncertainties of < 3 % at 2σ, comparable to the ~ 3.3 % (2σ) combined random, systematic, and standard uncertainty for HB-Line plutonium measurements [11].

For items which are not measured on the calorimeter, but still undergoing a known-α adjustment, the same -0.78 % bias is first applied. The total uncertainty is then calculated from a summing in quadrature of the uncertainties in the item measured on the calorimeter, α_C, and the measured singles rate. This is expressed in Equation 16.

16)
$$\sigma_{total}^2 = \sigma_{total\, i_1}^2 + \sigma_{\alpha_C}^2 + \sigma_S^2$$

Due to the relatively small uncertainties in the adjusted α value and the singles rate, typical adjusted mass uncertainties are ~ 3.0 % in the KIS process. A full assessment has not been made of typical uncertainty limits outside of the KIS process.

It would not be inaccurate to include uncertainties in the leakage multiplication and the spontaneous fission rates in the calculation of the total plutonium uncertainty. However, as these uncertainties are normally << 1 %, their inclusion was shown to have no significant impact on the total measured uncertainty. A similar statement cannot always be made about the inclusion of uncertainties in ε, f_d, or f_t, which relate directly to instrument calibration.

Application of Known-α Analysis

The known-α analysis technique has been evaluated and/or applied numerous times at SRS. In each application, a single plutonium (or mixed U/Pu) bearing item is divided into multiple containers. An item from that batch is then selected for both neutron and calorimetric measurements; all remaining items undergo a neutron measurement only (it may or may not be necessary to perform multiple in-house isotopic measurements). Using the described technique, the alpha value α_C determined from the first (calorimeter and passive neutron measured) item is used for all other items in the batch. The central motivation in each case is to achieve the required accuracy while minimizing measurement time.

The K-Area Interim Surveillance (KIS) Process

The K-Area Interim Surveillance (KIS) Facility is tasked primarily with support of the Department of Energy (DOE) 3013-Surveillance Program. This involves multiple nonnuclear measurements (gas sampling, radiography, etc.) in addition to nuclear measurements. The nuclear measurements require a suite of gamma ray, neutron, and heat detection instruments [12,13,14,15]. Thus, typical item surveillance is a multistep process involving both nondestructive and destructive aspects [16].

As part of the nondestructive examination (NDE) of a 3013 containing plutonium or a mixture of plutonium and uranium, the item undergoes gamma isotopic and neutron emission rate measurements. The combined results of these measurements then yield plutonium content. Similar results can also be determined for the uranium content, though this is a more indirect measurement depending on the uranium/plutonium ratio and declared enrichment. When required, these measured values are compared for agreement (within statistical uncertainty) with the expected masses.

Nuclear measurements for the NDE portion of a 3013-Surveillance typically require only the KIS neutron multiplicity counter (KIS-NMC) and Gamma Isotopic System (KIS-GIS). The results of these measurements then determine the path forward for any daughters produced in the DE process. Specifically, in accordance with DOE performance requirements, when the uncertainty in the plutonium content is no greater than 7.0 % (or 12.5 % for the ^{235}U uncertainty), it is sufficient to assay the daughter cans using only a gamma-ray/neutron combination, similar to that used for the original 3013. However, when uncertainties exceed these thresholds, a calorimeter measurement is required for one of the daughters. The results of this additional measurement are then used in determining the plutonium masses for the remaining daughters. Application of this method in the current KIS scheme has been shown to produce uncertainties ~ 3 %.

Shuffler Equivalence to the KIS Process

The current nuclear measurement scheme for the 3013-Surveillance Program is limited to use of the KIS calorimeter (KIS-CAL), KIS-GIS and KIS-NMC, in addition to the portable high purity germanium detectors used in holdup and waste assay [14, 15]. Though the NMC also has active capability, useful in quantifying ^{235}U, its use has been limited to the passive assay of plutonium. Due to certain nuclear criticality safety requirements, which limit allowable material and overall operational efficiency, it has been proposed that the KIS-NMC be removed from the KIS Facility and thus, from the current 3013-Surveillance nuclear measurement scheme. While this removal would allow a relaxation of material and throughput restrictions, the prohibitively long (~ 8 hours) measurement time required by the KIS-CAL would still present a significant impact to operations, particularly when multiple assays are required.

Should the KIS-NMC be removed, the most feasible option is to use the K-Area ^{252}Cf Shuffler (Shuffler) as a passive multiplicity counter. As the instrument has undergone a multiplicity characterization, it would be a straightforward process to substitute the Shuffler into the current KIS measurement scheme [2]. The only consideration in making this substitution is the Shuffler's efficiency (~ 20 %), which is far less than the ~ 50 % efficiency of the KIS-NMC. This lower efficiency directly translates to the higher uncertainties observed in the Shuffler PuO_2 qualification results [17,18]. A comparison of the Shuffler and KIS-NMC multiplicity characterization parameters is given in Reference 5. As these parameters yield uncertainties which exceed the above mentioned Material Control and Accountability limits, all measured mass values would require calorimeter-based adjustment.

To validate application of the known-α analysis method to Shuffler measurements, a feasibility study was conducted. In this study, a series of items was selected which had previously run through both neutron and calorimeter measurements as part of the KIS DE process. The items underwent passive one-hour Shuffler measurements, consistent with the one-hour passive measurement scheme used in KIS.

The percent differences between the KIS values and the calorimeter-adjusted Shuffler plutonium results are illustrated in Figure 1. For comparison, the raw (unadjusted) results for the same items are also shown. From Figure 1 it is apparent that the lower counting efficiency of the Shuffler (likely coupled with impurities in certain items) results in plutonium values which greatly differ from the expected masses. Conversely, though the neutron instruments used in each case vary, use of known-α analysis has been shown to bring the measured results to within < 1.0 % of expectation.

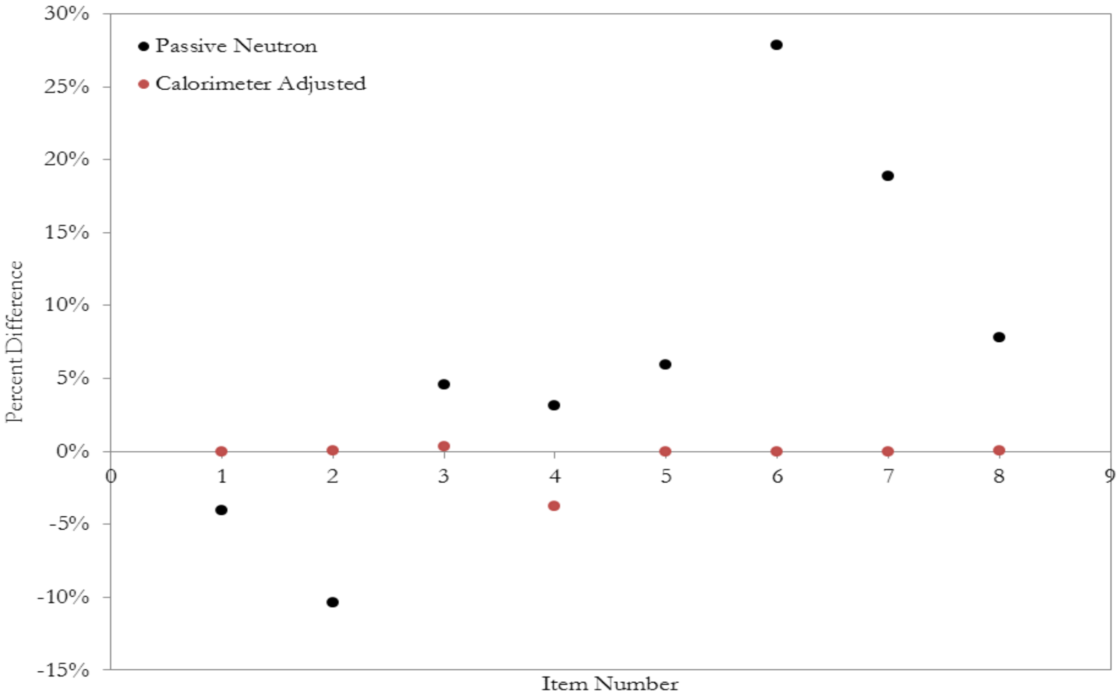

Figure 1 Comparison of passive neutron and known-α adjusted Shuffler assay results with established KIS plutonium mass values.

The H-Area Pu-Blending Campaign: Ship to WIPP (SW)

The purpose of the H-Area B-Line (HB-Line) Facility is the chemical dissolution and processing of nuclear material. This may be for a variety of reasons, including preparation of material for nuclear fuel production. HB-Line also served as the primary disposal path for all material passed through the KIS Process. Recently, a decision was made to stop all chemical processing in the HB-Line Facility. The result was a significant interruption to the pre-existing path to move plutonium from an accountable to unaccountable material status. An alternative path was therefore developed to continue processing the material in its solid form. Rather than dissolution, each KIS item is now divided into multiple containers and blended with what is termed termination of safeguards material. This blended material is then packaged and sent to the Waste Isolation Pilot Plant (WIPP) for burial.

Before the blended material can be shipped, it must be measured, with the results compared for agreement with the material mass input. The measurement instrument selected was the HB-Line active well coincidence counter HBL-AWCC. Before use in the SW campaign, the HBL-AWCC underwent a multiplicity characterization, qualifying it as a neutron multiplicity counter [19]. This characterization would allow use of the instrument for SW without undergoing a coincidence calibration, which would require representative standards and/or the extensive use of simulation.

Because this is a relatively low efficiency instrument (~ 20 %), mass results of the initial qualification had significantly high uncertainties, resulting in random and calibration uncertainties of 7.8 % and 12.9 % respectively [20]. To offset the limitations revealed in the instrument qualification, it was decided that known-α analysis be employed in this campaign. This method was also selected because a low efficiency instrument is especially susceptible to impurity effects.

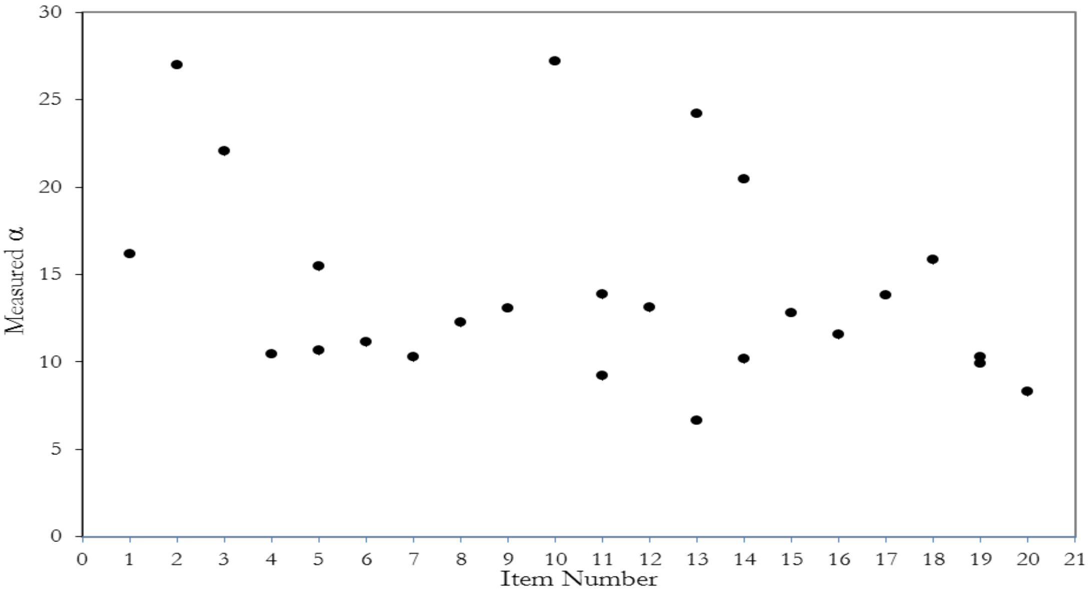

Figure 2 Measured α-values for a batch of Ship to WIPP samples with measurable impurities

Possible results of this susceptibility are displayed in Figure 2. For the data in Figure 2, a single item was split and blended into twenty (20) containers [21]. The items were then measured individually using the HB-Line NMC. Because these items have a common source, it is reasonable to expect identical (α,n):spontaneous fission ratios across the batch. However, it is clear that these items, which had measurable amounts of several impurities, presented a challenge to conventional multiplicity assay.

The effects of a large variability in the calculated α-value are clear from Figure 3, which compares the unadjusted (Pu neutron) results with the expected mass values. At times, the neutron-based mass values deviated by as much as 75 % from expectation. Conversely, through the use of calorimeter-based known-α multiplicity analysis, measured mass values align almost exactly with the declared values. The average percent difference between the expected and adjusted mass values for this item was 2.9 %, compared with 13 % for the unadjusted measurements.

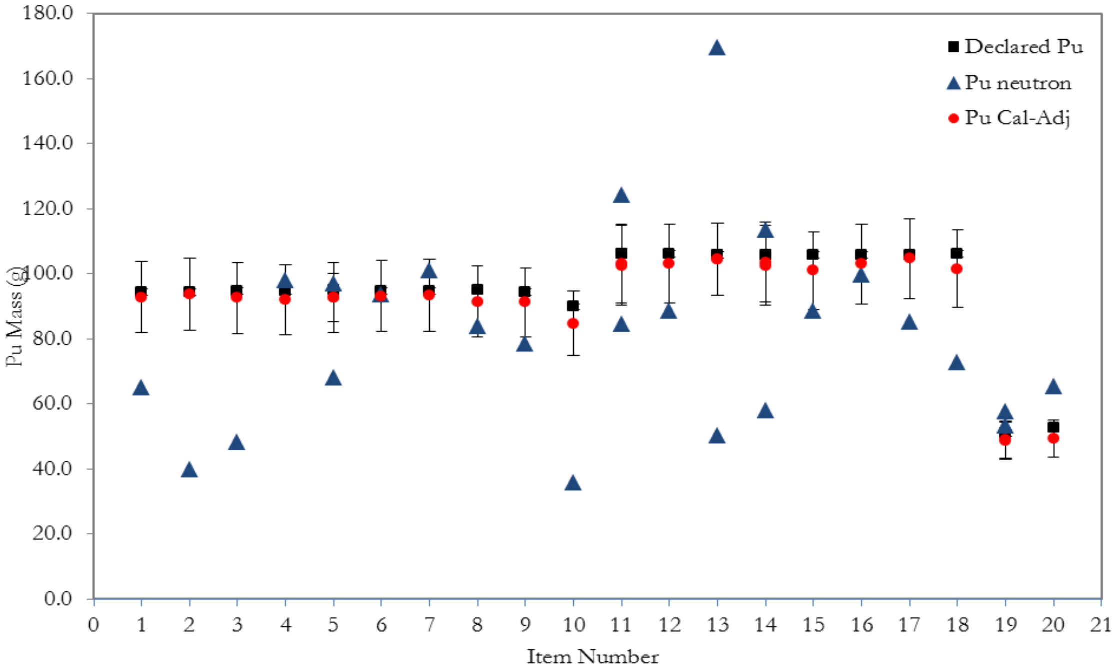

Figure 3 Comparison of unadjusted and calorimeter-adjusted H-Area plutonium masses with known KIS values. For visual clarity, uncertainties in the unadjusted mass values are omitted.

Conclusion

The precision and accuracy of passive neutron measurements are largely driven by counting statistics. Consequently, plutonium measurements which use the common gamma-ray/passive neutron combination can be highly sensitive to the presence of impurities, particularly (α,n) emitters. As a result of this sensitivity, neutron measurements of material containing measurable impurities can result in higher uncertainties and/or skewed mass results. Thus, the time gains of passive measurements are offset by a loss in accuracy. Conversely, a scheme which uses a combination of gamma ray and calorimetric measurements has been shown to give much higher accuracy than typical neutron measurements. This is owed to the reliance of the calorimetric measurements on bulk material heat output signatures, rather than statistical analysis. However, the approximately 8+ hour minimum time requirement of calorimeter measurements makes this the least preferred method when high throughput is desired.

The calorimeter-based known-α multiplicity analysis scheme was therefore developed as a means to combine the accuracy and precision of a calorimetric measurement with the relative speed of passive neutron counting. It is important to note that the method is uniquely suited to batch analysis, i.e. measurements of multiple plutonium items having a common source. This is because the method requires gamma, neutron, and heat measurements of a representative sample from the batch; combination of these results is used to determine α, the (α,n):spontaneous fission neutron ratio. Based on an assumption of homogeneous distribution of source material, other items in the batch then only require passive neutron measurements. The passive multiplicity analysis performed for each subsequent item then incorporates the previously determined α value.

Use of a calorimetric measurement to determine α as an input to the conventional multiplicity equations has no instrument or mass limitations. Thus, the KIS calorimeter has been shown to produce viable inputs to multiplicity analysis in the high-efficiency (50 %) KIS-NMC, as well as the much lower efficiency (~ 20%) K-Area Shuffler. The method has also been applied, with no alteration, using H-Area instruments for the Pu-Blending campaign, which packages material in lower quantities and in a vastly different matrix than that used in KIS. Further, while no specific examples are mentioned herein, uniformity of batch-item mass is not required. Thus, the α-value determined from calorimetric measurements of a relatively large batch item, or from the source material itself, can be used in known-α analysis of subsequent batch items.

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