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Known-Multiplication Neutron Assays for Dilute Surplus Plutonium

Timothy Aucott, Christopher McGrath
Savannah River National Laboratory

Abstract

Neutron coincidence counting was chosen as the preferred method for assay of diluted surplus plutonium in Criticality Control Overpacks (CCOs) produced at the Savannah River Site. However, a significant 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 erroneously high readings when analyzed with a standard calibration curve. These biases can result in measurements falling outside required bounds for both Material Control & Accountability and Waste Acceptance Criteria.

A known-multiplication analysis has been implemented for verification measurements on these items. This method assumes that the multiplication can be predicted for a given item based solely on its isotopic ratios. This allows for the singles and doubles coincidence rates to be solved for the plutonium mass and (α, n) emission rate. Due to the consistent geometry and composition of each CCO, the multiplication is particularly well behaved across a range of isotopics, mass loadings, and impurity contents. Calibration standards have been made to empirically determine the multiplication in the CCOs, and working standards have been created to determine the accuracy and precision of the method.

Introduction

As part of the Dilute and Dispose work currently underway at the Savannah River Site (SRS), excess plutonium materials will be diluted and discarded to the Waste Isolation Pilot Plant (WIPP). Dilute plutonium-bearing items will be packaged into criticality control overpack (CCO) containers. CCOs will be assayed to verify plutonium content for Nuclear Material Control and Accountability (NMC&A), as well as to ensure compliance with WIPP Waste Acceptance Criteria (WAC). Neutron coincidence counting was chosen as the preferred method for assay of the material. Two high efficiency neutron counters (HENCs) [1][2][3] will be used to measure the effective ^{240}Pu content of each CCO.

The majority of the material to be dispositioned consists of relatively pure PuO_2, which will be easily assayed by the HENCs. However, the remainder consists of scrap oxide generated at various sites across the DOE complex. These impure oxides contain high quantities of light elements (in particular, those with atomic numbers less than 18), which will result in erroneously high readings on the HENCs [4][5][6].

To address the anticipated uncertainties, the plutonium inventory has been divided into measurement families and sub-families, based on the relative amounts of various impurities. The characteristics of each family and subfamily are chosen based on their predicted bias when the material is assayed on the HENC. The families are defined by the relative amount of (alpha, n) neutrons compared to spontaneous fission (referred to as "alpha" in neutron counting), and sub-families are defined by the dominant impurity type (e.g., beryllium, fluorine, etc.).

Calibration standards have been made using the same dilution process and characterized by thermal ionization mass spectrometry (TIMS) and calorimetry. These calibration standards have been used to calibration the HENCs and demonstrate the effectiveness of the known-multiplication method. Working standards, representing each family and sub-family, will also be created over the course of the campaign and characterized by gamma spectroscopy and calorimetry. These working standards are used to verify the accuracy and precision of the method across the range of impurities.

A single calibration was performed that encompassed three different analysis methods: coincidence counting, known-multiplication, and multiplicity. While the coincidence counting and multiplicity analyses were unchanged from the usual implementation, the known-multiplication approach was adjusted to match the specific matrix of interest.

Method Description

In the known-multiplication approach [7][8][9][10], both the singles and the doubles are used in order to improve the accuracy of the HENC results. This assumes that the multiplication M is well-behaved, but that the $^{240}Pu_{eff}$ mass m and the alpha ratio α are unknown. In the same way that traditional neutron counting uses the $^{240}Pu_{eff}$ wt%, this approach also uses the $^{239}Pu_{eff}$ wt% to determine the sample multiplication. Both singles and doubles rates are assumed to be background corrected.

The expected singles S and doubles D for an item assayed on a given neutron counter is as follows:

$$S = m\, F_0\, \varepsilon\, \nu_{s1}\, M\, (1+\alpha) \tag{1}$$

$$D = \tfrac{1}{2} m\, F_0\, \varepsilon^2\, f_D\, \nu_{s2}\, M^2\, [1 + k_\alpha (M-1)(1+\alpha)] \tag{2}$$

The efficiency ε and doubles gate fraction f_D are calibration constants specific to the instrument, and F_0, ν_{s1}, ν_{s2}, and k_α are physical constants [11]. For simplicity, two instrument-specific constants, S_0 and D_0, are defined as follows:

$$S_0 = F_0\, \varepsilon\, \nu_{s1} \tag{3}$$

$$D_0 = \tfrac{1}{2}\, F_0\, \varepsilon^2\, f_D\, \nu_{s2} \tag{4}$$

For the known-multiplication approach, it is assumed that the multiplication can be expanded as follows:

$$M = 1 + b_M \frac{m}{k_M} + \cdots \tag{5}$$

where b_M is a calibration constant specific to the sample matrix, and k_M is the $^{240}Pu_{eff}$ to $^{239}Pu_{eff}$ ratio for a given item. For this particular material, the multiplication is expected to be low enough that any higher order terms are negligible. However, higher order terms could be included for other applications with higher sample multiplication.

In order to determine the mass of an unknown item, the equations are solved as follows. The ratio k_M must first be known for the unknown item. Next, alpha can be eliminated from the two equations, giving the following quadratic expression:

$$0 = -\left[\frac{D}{D_0}\right] + m\left[1 + k_\alpha \left(\frac{b_M}{k_M}\right)\left(\frac{S}{S_0}\right)\right] + m^2\left[2\left(\frac{b_M}{k_M}\right) + k_\alpha \left(\frac{b_M}{k_M}\right)^2 \left(\frac{S}{S_0}\right)\right] \tag{6}$$

which is solved by the quadratic equation to determine the ^{240}Pu effective mass m.

Calibration Approach

Standard Creation

Calibration standards were made using well characterized Alternate Feedstock (AFS) oxide, item ID CCH6262, originally produced in HB-Line. The plutonium isotopics were characterized at F/H Analytical Laboratories by TIMS in May 2018. CCH6262 was downblended in the KIS glovebox to create 10 blend can standards, each with a prescribed net weight of oxide.

After downblend, each blend can standard was assayed in triplicate on one of the KIS calorimeters. The three runs were averaged together to determine the reported power. Specific power from TIMS, along with the power from calorimetry, was used to determine the plutonium content. Matching pairs of blend can standards were then loaded into CCOs to create the CCO calibration standards.

Calibration Procedure

The calibration parameters for the HENCs were determined in such a way as to give unbiased results for a range of analysis techniques. The calibration approach described here is consistent with that outlined in the Application Guide to Neutron Multiplicity Counting [4], with some small additions to provide consistency with the doubles-based analysis techniques.

First, the doubles calibration curve is determined using a standard least-squares fit, as shown in Figure 1. The resulting equation for doubles D as a function of effective ^{240}Pu mass m_{240} is:

$$D = 57.07\, m_{240} + 0.507\, m_{240}^2 \qquad (7)$$

This provides the calibration for the standard doubles counting method, as well as the doubles response D_0, equal to 57.07 counts per second.

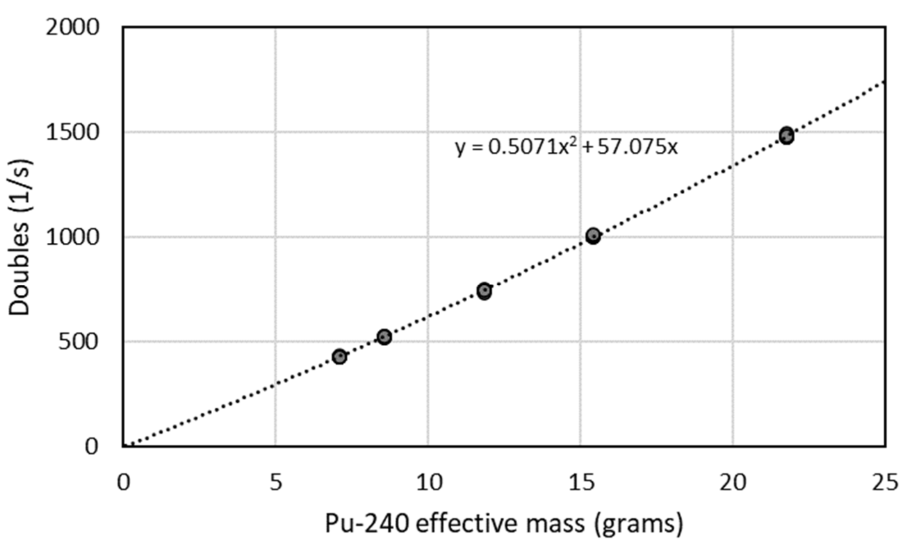

Figure 1: Doubles calibration curve for the calibration standards

Next, the doubles response is used to calculate the overall counting efficiency. The nominal efficiency is 0.3067, as determined using a range of ^{252}Cf sources at the factory. However, the efficiency for a CCO is expected to be somewhat larger due to scattering in the sample and drum components [12]. In contrast, the doubles gate fraction f_D is expected to remain consistent between the ^{252}Cf sources and the CCO. The nominal doubles gate fraction of 0.6267 was left unchanged, while the efficiency ε and singles response S_0 can be calculated as follows:

$$\varepsilon = \sqrt{\frac{2\,D_0}{F_0\, f_D\, \nu_{s2}}} = 0.3186 \tag{8}$$

$$S_0 = F_0\, \varepsilon\, \nu_{s1} = 325.0 \tag{9}$$

For the multiplicity analysis, the triples gate fraction f_T was adjusted to 0.4393 to minimize the root-mean-squared error in the measured ^{240}Pu mass. This represents only a 1.3% change from the factory-measured value for ^{252}Cf.

Finally, to calibrate the known-multiplication approach, the relationship between the fissile plutonium mass and multiplication is required. The slope of this line b_M can be determined by fitting the measured multiplication as a function of effective ^{239}Pu mass. The multiplication can be determined by standard multiplicity analysis, or (because the true plutonium mass of the standards is well known) by solving the singles and doubles equations for multiplication and alpha. The multiplication results for both approaches are shown in Figure 2, showing excellent agreement between the two analyses. The least-squares fit line to these data gives a value of 7.52×10^{-5} for b_M.

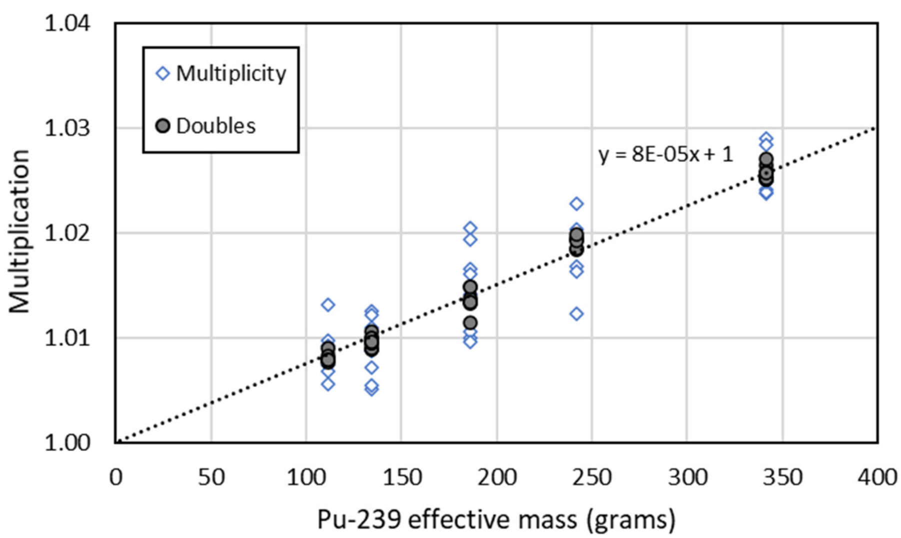

Figure 2: Multiplication as a function of effective ^{239}Pu mass. Measured values are shown for multiplicity analysis (blue diamonds) and doubles analysis (black circles).

Results

At the time of writing, three material families have been measured on the SRS HENCs: pure oxide (defined as $\alpha<3$), fluorine-bearing oxides, and beryllium-bearing oxides. Figure 3 compares the measured doubles rate to the calibration curve for all items measured to date. For the impure plutonium items, the light element will produce excess neutrons, which in turn induce additional fissions in the plutonium and bias the HENC assay high.

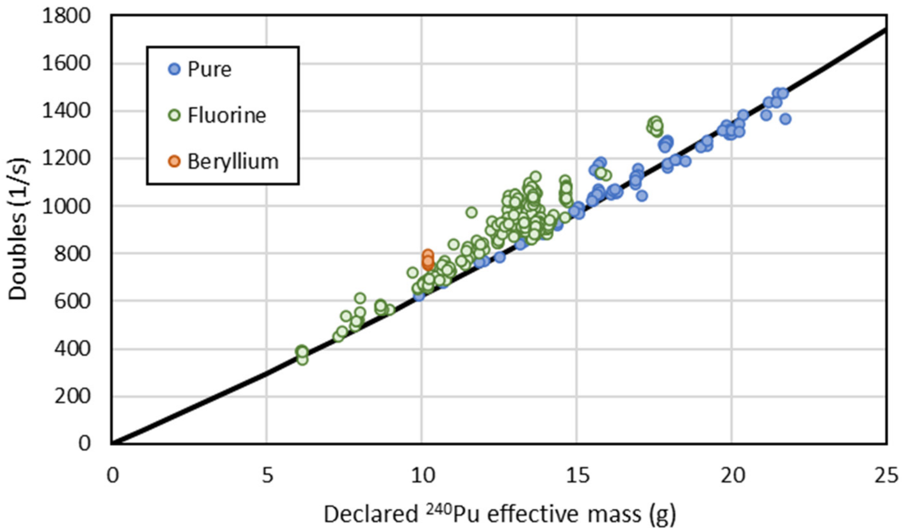

Figure 3: Measured doubles rate as a function of declared effective ^{239}Pu mass. The solid line shows the calibration curve, while the data points show results for pure (blue), fluorine-bearing (green), and beryllium-bearing (orange) oxides.

Although the calibration curve does not explicitly calculate alpha, the singles to doubles ratio provides a reasonable estimate of the level of impurities in these drums. Figure 4 shows the positive bias which increases with impurity level, consistent with the predicted effect.

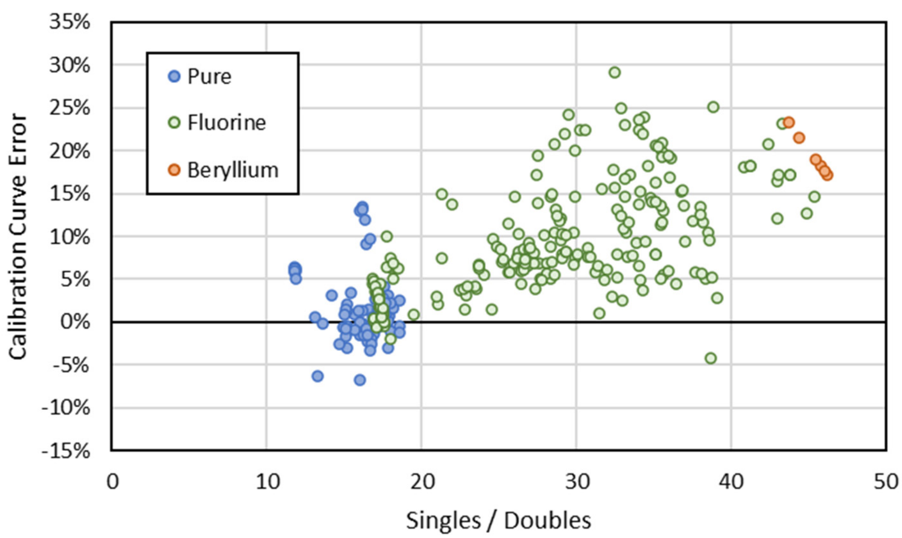

Figure 4: Error between declared and measured effective ^{239}Pu mass by calibration curve. Data points show results for pure (blue), fluorine-bearing (green), and beryllium-bearing (orange) oxides.

The known-multiplication error should, in principle, have a negative bias fluorine-bearing items, and a positive bias for beryllium-bearing items, due to the difference in energy of the corresponding (α,n) neutrons. While the average bias was slightly negative for fluorine, the bias was not significantly different from zero. For beryllium, only a single working standard has been created to date, so the number of samples in insufficient to determine if the bias is significant. Figure 5 shows the known-multiplication error as a function of alpha.

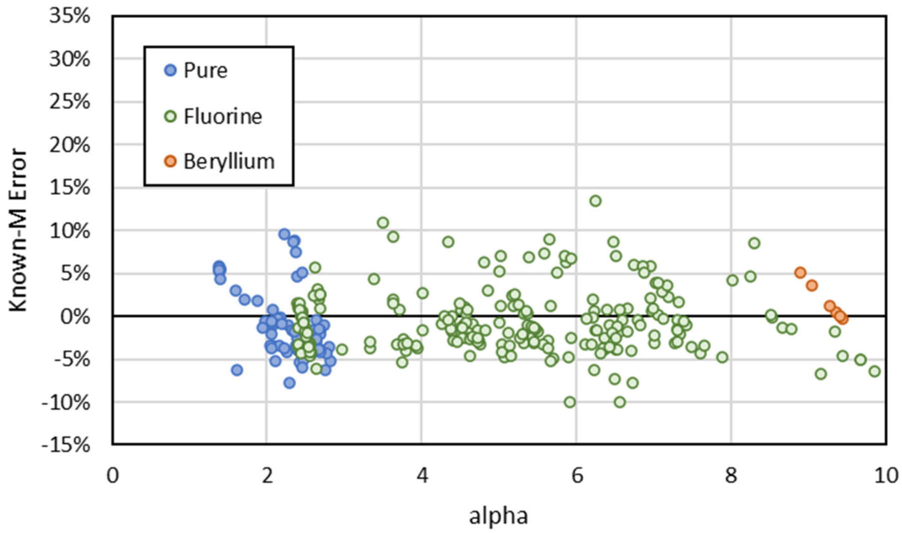

Figure 5: Error between declared and measured effective ^{239}Pu mass by known-multiplication. Data points show results for pure (blue), fluorine-bearing (green), and beryllium-bearing (orange) oxides.

Finally, multiplicity was also evaluated for these items. The HENCs were not designed as multiplicity counters, and alpha is high enough for these items that multiplicity analysis has a prohibitively large random uncertainty (due to the low precision in the triples rate). Figure 6 shows that the multiplicity analysis is overall unbiased, but that the random uncertainty increases with alpha, as expected.

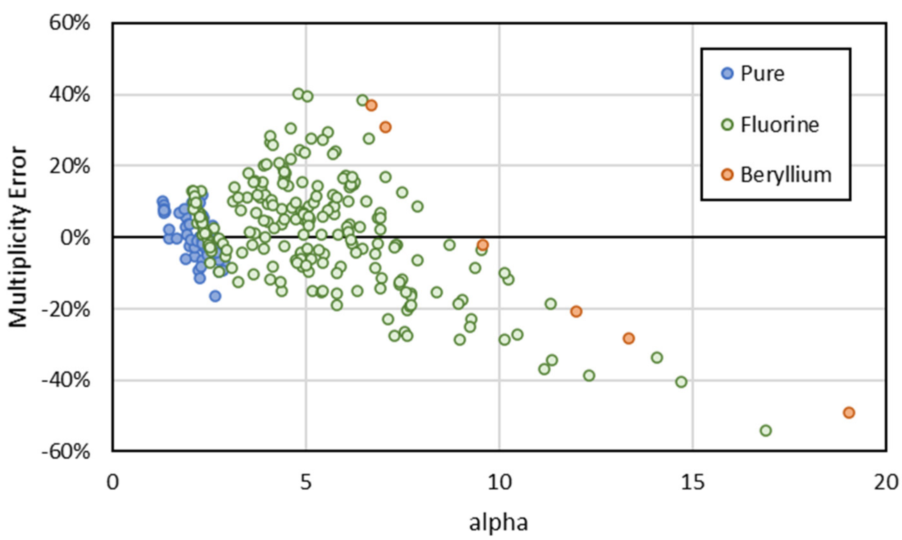

Figure 6: Error between declared and measured effective ^{239}Pu mass by multiplicity. Data points show results for pure (blue), fluorine-bearing (green), and beryllium-bearing (orange) oxides.

Conclusions

The known-multiplication method has been used for verification of plutonium content in diluted CCOs. Because of the dilute matrix and consistent geometry, this method is able to rely on a linear relationship between the effective ^{239}Pu content and the sample multiplication. Two HENCs were calibrated to allow for calibration curve, known-multiplication, and multiplicity analyses with the same calibration constants.

The standard doubles calibration curve shows the expected positive bias which increases with matrix impurity. In some applications, this positive bias may provide an acceptable conservativism. However, the known-multiplication analysis successfully corrects for this bias.

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