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# Systematic Effects in Neutron Coincidence and Multiplicity Counting

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Correlated neutron counting, including neutron coincidence and multiplicity counting, is an important tool in nuclear material accountancy verification. The accuracy of such measurements is of interest to the safeguards community because as the accuracy of NDA improves, the number of samples that are required to undergo destructive analysis (DA) decreases. The accuracy of a neutron multiplicity measurement can be affected by a number of variables. Monte Carlo neutron transport simulations with MCNPX have been performed to understand how the properties of the sample affect the count rate. These resultant count rates have been analyzed with the "point model" in order to determine the effect on the deduced plutonium mass. The sample properties that have been investigated are density, sample position within the detector cavity, moisture content, isotopic composition, plutonium to total actinide ratio and heavy metal fraction. These parameters affect the Singles, Doubles and Triples count rates in different ways. In addition, different analysis methods use these measured quantities in different combinations, so that the final sensitivity of the $^{240}$Pu mass to each parameter also depends on the analysis method used. For example, the passive calibration curve method only used the Doubles rate to produce the $^{240}$Pu mass and so is not sensitive to changes in the Singles rate (to first order). The analysis methods considered here were passive calibration curve (non-multiplication corrected), known alpha (multiplication corrected) and multiplicity with known efficiency. The effects were studied on both a small mass MOX sample (1 g Pu) and a large MOX sample (6000 g Pu) both measured in high efficiency neutron multiplicity counters. In order to determine the final effect of each parameter it is necessary to know not only the sensitivity of the plutonium mass to that parameter, but also the range over which the parameter can realistically vary. Some estimates are given.

## Introduction

Correlated neutron counting, including neutron coincidence and multiplicity counting, is commonly used at nuclear fuel cycle facilities to non-destructively assay items containing plutonium (Pu). The results of a passive neutron assay can be expressed as an effective mass of $^{240}$Pu, $^{240}$Pu$_{\text{eff}}$, given by equation 1 [1]:

$$^{240}Pu_{eff} = 2.52 f_{238} + f_{240} + 1.68 f_{242} \qquad \text{(Equation 1)}$$

Where, $f_i$ is the mass (or mass fraction) of the spontaneously fissile plutonium isotope $i$. The mass (or mass fraction) of each isotope is multiplied by a coefficient which weights the contribution of that isotope to the $^{240}$Pu effective mass [2]. The total Pu mass can be derived from the $^{240}$Pu effective mass, given known isotopic composition.

Neutron count rates (Singles, Doubles and Triples) depend on the mass of spontaneously fissile material present, characteristics of the detection system (e.g. efficiency as a function of sample position and energy), and several assay parameters including the alpha ratio and net leakage

multiplication. Alpha, $\alpha$, is the ratio of the neutron production via ($\alpha$, n) reactions to the neutron production via spontaneous fission. Systematic variations in sample properties, such as the six studied here, can alter the assay parameters and thus lead to uncertainties in the measured counting rates and hence measured mass. Uncertainties also arise from the chosen analysis method because each analysis method uses a different combination of count rates and assay parameters.

The accuracy of the final assay result largely depends on the accuracy of the system calibration; which should be improved by using reference samples that closely represent the characteristics of the final items being assayed. The accuracy target of a high efficiency neutron multiplicity counter is < 0.5%. However, a number of unknown or poorly characterized sample parameters can limit the accuracy with which the total Pu mass can be determined. Thus, the goal here is to understand the effect of a range of system parameters on the derived Pu mass and furthermore how systematic uncertainties propagate in the following analysis methods: passive calibration curve ("non-multiplication corrected"), known alpha ("multiplication corrected"), and multiplicity with known efficiency.

In this work, we simulate the detector response of two neutron multiplicity counters and investigate the effect of small perturbations of a variety of sample parameters on the resultant "measured" plutonium (Pu) mass, derived from both standard coincidence counting and multiplicity analyses. Using this approach, we can better understand how the properties of a MOX sample affect the results of both neutron coincidence and multiplicity counting. We can then infer the importance of the treatment of each parameter in the different analyses and whether additional correction factors will need to be researched in the future.

## Model

Monte Carlo neutron transport simulations with MCNPX (version 2.7.A) [3] have been performed to determine the effect of a range of sample properties on the detector response (Singles, Doubles and Triples count rates). The measurement of a small mass mixed oxide (MOX) sample (1 g Pu) and a large MOX sample (6000 g Pu) were simulated. Two high efficiency neutron multiplicity counters were modeled to simulate the correlated neutron measurements of these samples: (1) the Epithermal Neutron Multiplicity Counter (ENMC) [4], in the case of the large MOX sample; and (2) a high efficiency inventory sample counter ("ISC"), in the case of the small MOX sample. Both detectors are fielded for MOX assay applications.

## Sample Parameter Perturbations

The following sample parameters were varied (25 sample perturbations): bulk density, sample position within the detector cavity, moisture content, the presence of $^{241}$Am from Pu decay, plutonium ratio (mass of plutonium relative to total actinide mass), and heavy metal fraction relative to oxide (i.e. $XO_2$ vs. $X_3O_8$, where X is Pu + U). A further 6 sample perturbations were modeled by varying the plutonium and uranium isotopic composition. A total of 32 cases were modeled: 1 reference sample and 31 perturbations. Table 1 details the first 26 cases including the reference values of these sample parameters and the simulated ranges over which these parameters were varied.

Table 1. Sample parameter reference values, sample perturbation values and simulated range of perturbations

| Sample Parameter | Reference Value | Sample (Perturbation) | Parameter Value (ISC/ ENMC) | Simulated Range (ISC) | Simulated Range (ENMC) |
|---|---|---|---|---|---|
| Density | 2.3 g/cm$^3$ | 1 | 1.5 g/cm$^3$ | | |
| | | 2 | 1.7 g/cm$^3$ | | |
| | | 3 | 1.9 g/cm$^3$ | | |
| | | 4 | 2.1 g/cm$^3$ | | |
| | | 5 | 2.5 g/cm$^3$ | | |
| | | 6 | 2.7 g/cm$^3$ | | |
| | | 7 | 2.9 g/cm$^3$ | | |
| | | 8 | 3.1 g/cm$^3$ | | |
| | | 9 | 3.3 g/cm$^3$ | | |
| | | 10 | 3.5 g/cm$^3$ | 1.5 to 3.5 g/cm$^3$ | 1.5 to 3.5 g/cm$^3$ |
| x-position | Center (0) | 11 | 0.7 cm/ 1.0 cm | 0 to 0.7 cm | 0 to 1.0 cm |
| y-position | Center (0) | 12 | -0.7 cm/ -1.0 cm | -0.7 to 0.7 cm | -1.0 to 1.0 cm |
| | | 13 | 0.7cm / 1.0 cm | | |
| Moisture content | 0 % | 14 | 1 % | 0 % to 4 % | 0 % to 4 % |
| | | 15 | 2 % | | |
| | | 16 | 3 % | | |
| | | 17 | 4 % | | |
| Pu ratio[b] | 0.5 | 18 | 0.07121/ 0.3073 | 0.07121 to 1.0 | 0.3073 to 1.0 |
| | | 19 | 0.2/ 0.35 | | |
| | | 20 | 0.4 | | |
| | | 21 | 0.6 | | |
| | | 22 | 0.8 | | |
| | | 23 | 1.0 | | |
| Metal fraction | 0.8815 | 24 | 0.848 | 0.848, 0.8815 (discrete) | 0.848, 0.8815 (discrete) |
| $^{241}$Am presence | No | 25 | Yes | No, Yes (discrete) | No, Yes (discrete) |

[a] *Pu ratio is given by (total Pu mass)/(total Pu mass + total U mass)*

[b] *Metal fraction is given by (total Pu mass + total U mass)/(total sample mass). The reference material is $XO_2$ and the perturbed material is $X_3O_8$, where X is Pu + U.*

Note that the parameters were independently varied, but the mass of plutonium (1g or 6000 g) was preserved in each case. For the reference case and each perturbed case, one Monte Carlo calculation was run with an (α, n) neutron source and one with a spontaneous fission source, in order to compute the Singles, Doubles and Triples count rates (for both neutron sources and all detector channels). The measured plutonium mass was then calculated from the resultant count rates using the following analysis methods: standard calibration curve ("non-multiplication corrected"), known alpha ("multiplication corrected"), and multiplicity with known efficiency. Results are presented for the first 26 cases in the following section. Results for variations in isotopic composition are considered separately.

## Results

The change in the measured mass is defined as the difference between the measured mass ("inspector measured mass") and the "operator declared mass" (i.e. the "best guess" of the true mass of the sample). The mass of the reference sample was defined to be the same as the operator declared mass, so that the change in the measured mass from the declared mass was zero in the case of the reference sample. Effects of the sample parameters on the mass were then defined relative to the reference sample.

### Estimated Parameter Ranges

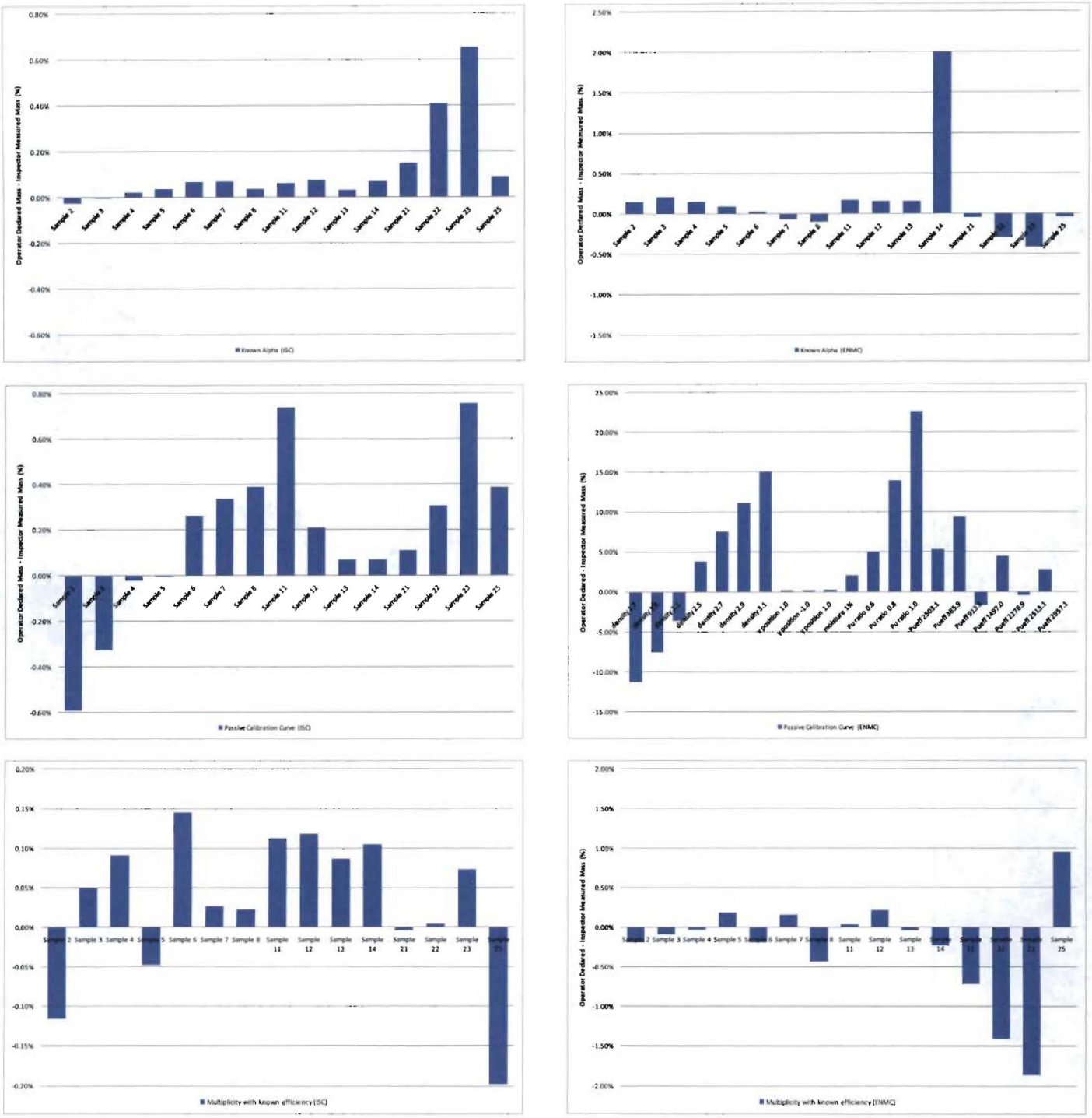
In order to determine the final effect of each parameter it is necessary to know not only the sensitivity of the plutonium mass to that parameter, but also the range over which the parameter can realistically vary. The range over which each sample parameters can vary has been estimated, based on our experience. Firstly, the sensitivity to each parameter was calculated over a range of values that were estimated to cover the complete range of interest. The realistic range for particular applications was also considered. Thus, the change in the measured mass is presented for two ranges of sample parameters in some cases.

### Dependence of Sensitivity on Sample Parameter

Figure 1 provides a visual representation of the effect of each sample parameter on the measured mass. Results are presented for both the small MOX sample (ISC) and the large MOX sample (ENMC). The first major point is that all of the parameters affect the large MOX sample to a greater extent than the small MOX sample. In the non-multiplication corrected case, the results are significantly affected by parameters that change the multiplication (i.e. density, moisture, Pu ratio) whereas for the multiplication corrected case, the largest change is caused by an "incorrect" estimate of the alpha value. Alpha is the ratio of the neutron production via (alpha, n) reactions to the neutron production via spontaneous fission. In order to apply the known alpha analysis method (thus apply multiplication correction) a "dry" value for alpha is assumed in the calculation of $^{240}Pu_{eff}$. In reality this would change with e.g. moisture content, but this value is not known prior to an assay.

Results tables 2 – 7 are provided for each sample property. Overall, the known alpha analysis method appears to be most sensitive to changes in metal fraction – this could be symptomatic of our assumptions i.e. certain oxide fraction. This increases the Totals rate, but doesn't strongly influence the Doubles. The standard calibration method is most sensitive to changes in the Pu ratio.

Figure 1. Change in mass as a function of sample parameter perturbation, for the first 25 perturbations (see table 1)



Density

Changes in density were considered over the full simulated range of perturbations (see table 1) and a smaller, more realistic range.

Table 2. Change in mass (%) for the three analysis methods as a function of changes in density

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 1 – 10 | 1.5 to 3.5 g/cm$^3$ | 0.16 | 1.40 | 0.16 |
| ISC | | 2.03 to 2.39 g/cm$^3$ (2σ) | 0.04 | 0.25 | 0.04 |
| ENMC | 1 – 10 | 1.5 to 3.5 g/cm$^3$ | -0.23 | 37.83 | 0.02 |
| ENMC | | 2.03 to 2.39 g/cm$^3$ (2σ) | 0.07 | -6.73 | 0.14 |

Sample Position (x and y)

During a measurement, the MOX assay sample may not be placed centrally within the detector cavity thus the x and y sample positions may vary. Variations in position were considered over the full simulated range (see table 1).

Table 3. Change in mass (%) for the three analysis methods as a function of changes in sample position (x and y)

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 11 | 0.0 to 0.7 cm | 0.06 | 0.74 | 0.11 |
| ENMC | 11 | 0.0 to 1.0 cm | 0.17 | 0.16 | 0.03 |
| ISC | 12 - 13 | -0.7 to 0.7 cm | -0.04 | -0.14 | -0.03 |
| ENMC | 12 - 13 | -1.0 to 1.0 cm | 0.00 | 0.04 | -0.25 |

Moisture Content

Changes in sample moisture content were considered over the full simulated range (see table 1) and a smaller realistic range, based on our experience.

Table 4. Change in mass (%) for the three analysis methods as a function of changes in moisture content

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 14 - 17 | 0 to 4 % | 7.75 | 0.18 | 0.02 |
| ISC | | 0.15 to 0.23 % | 0.17 | 0.00 | 0.00 |
| ENMC | 14 - 17 | 0 to 4 % | 6.80 | 10.55 | -1.61 |
| ENMC | | 0.15 to 0.23 % | 0.14 | 0.21 | -0.03 |

MOX samples requiring verification measurements e.g. powders may absorb moisture from the atmosphere if left exposed [5]. Stewart, *et al.* [6] describes the three principal effects of moisture on the measured Singles (Totals) and Doubles (Reals) count rates from passive neutron coincidence verification measurements of $PuO_2$ and MOX powders: (1) the number of neutrons produced by (α, n) reactions is increased because water provides more oxygen nuclear targets for neutron production via α-particles. An increase in moisture therefore leads to an increase in Singles rate and an increase in the alpha ratio. (2) An increase in moisture leads to an increase in the number of neutrons produced by induced fission because average neutron energies are lowered in elastic collisions with hydrogen nuclei, thus sample moderation is increased. The detected Doubles (or Reals) rate is therefore increased. (3) Detection efficiency is increased in under-moderated counters due to the reduction in average neutron energy.

The magnitude of the effects of sample moisture content will also vary with sample isotopic composition due to the presence of isotopes that decay via α-particle emission i.e. the alpha particle yield will vary with sample isotopic composition, hence changing the neutron yield due to (α, n) reactions and therefore changing the alpha ratio e.g. $^{238}Pu$ is a major α-particle emitter and its presence could increase the impact of moisture on the alpha ratio.

Stewart, *et al.* [6] concluded that the primary contributor to moisture bias in the multiplication-corrected Reals assay of MOX samples studied is the change in α. Croft [5] quantified the enhancement of the (α, n) neutron production rates in a variety of MOX samples to make allowances for this effect.

Pu Ratio

Changes in the Pu ratio were considered over the full simulated range (see table 1) and a smaller realistic range, based on the two types of sample that would be measured during an assay: MOX with a Pu ratio of 0.5 and $PuO_2$ with a Pu ratio of 1.0.

Table 5. Change in mass (%) for the three analysis methods as a function of changes in Pu ratio

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 18 - 23 | 0.07121 to 1.0 | 1.18 | 1.75 | 0.13 |
| ISC | | 0.5 to 1.0 | 0.65 | 0.75 | 5.40 |
| ENMC | 18 - 23 | 0.3073 to 1.0 | -0.58 | 33.48 | -2.22 |
| ENMC | | 0.5 to 1.0 | -0.42 | 22.59 | -1.86 |

Metal Fraction

Changes in the sample metal fraction were considered over the full simulated range (see table 1).

Table 6. Change in mass (%) for the three analysis methods as a function of changes in metal fraction

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 24 | 0.8815 to 0.848 | 9.99 | 0.06 | -0.08 |
| ENMC | 24 | 0.8815 to 0.848 | 9.74 | 2.15 | 0.09 |

$^{241}$Am Presence

Two measurement scenarios were considered: with and without $^{241}$Am present.

Table 7. Change in mass (%) for the three analysis methods as a function of $^{241}$Am presence

| Detector | Perturbations | Change in Parameter | Change in Mass (%) Known alpha | Change in Mass (%) Passive calibration | Change in Mass (%) Multiplicity |
|---|---|---|---|---|---|
| ISC | 25 | No, Yes | 0.09 | 0.39 | -0.20 |
| ENMC | 25 | No, Yes | -0.04 | 5.30 | 0.95 |

**Isotopic Composition**

Table 8 shows the plutonium isotopic composition variations with the corresponding effective $^{240}$Pu mass, $^{240}Pu_{eff}$, expressed as a weight fraction (effective mass of $^{240}$Pu per gram of sample). For the

31 cases in which there was uranium in the sample (one case had none), the uranium isotopes (weight fraction) were $^{234}U$, 0.000051; $^{235}U$, 0.01; $^{236}U$, 0.005; and $^{238}U$, 0.984949.

Table 8. Plutonium Isotopic Composition (Weight Fraction)

| Isotope | Sample 26 | Sample 27 | Sample 28 | Sample 29 | Reference Sample | Sample 30 | Sample 31 |
|---|---|---|---|---|---|---|---|
| $^{238}Pu$ | 0.00011 | 0.00070 | 0.00846 | 0.01197 | 0.01271 | 0.01721 | 0.01975 |
| $^{239}Pu$ | 0.93412 | 0.84337 | 0.73319 | 0.62525 | 0.60375 | 0.58095 | 0.55476 |
| $^{240}Pu$ | 0.06313 | 0.14206 | 0.18295 | 0.25406 | 0.25469 | 0.24768 | 0.28438 |
| $^{241}Pu$ | 0.00223 | 0.01028 | 0.05463 | 0.06679 | 0.08334 | 0.09771 | 0.07155 |
| $^{242}Pu$ | 0.00040 | 0.00358 | 0.02077 | 0.04193 | 0.04551 | 0.05645 | 0.06956 |
| $^{240}Pu_{eff}$ [a] | 0.02835 | 0.06711 | 0.10997 | 0.1674 | 0.1722 | 0.1846 | 0.2172 |
| $^{240}Pu_{eff}$ (g) [b] (ISC) | 0.06432 | 0.1523 | 0.2495 | 0.3798 | 0.39073 | 0.4188 | 0.4929 |
| $^{240}Pu_{eff}$ (g) [b] (ENMC) | 385.9 | 913.6 | 1497. | 2279. | 2344. | 2513. | 2957. |

[a] *$^{240}Pu_{eff}$ is the effective $^{240}Pu$ mass, given by equation 1. This table shows $^{240}Pu_{eff}$ per gram of plutonium.*

[b] *The effective Pu mass of the MOX sample, $^{240}Pu_{eff}$ (g) was obtained by multiplying the sample $^{240}Pu_{eff}$ value as a weight fraction by the sample density and volume, which were the same for all variations.*

The variation in the measured mass as a function of changes in isotopic composition was considered as several separate cases of interest. A number of measurement scenarios are considered in table 9.

Table 9. Change in mass (%) for the two standard coincidence analysis methods as a function of isotopic composition

| Safeguards Measurement Scenario | Change in Isotopic Composition | Change in Mass (%) Multiplication Corrected | Change in Mass (%) Non-Multiplication Corrected |
|---|---|---|---|
| Full reference range (greatest change in $^{239}Pu$) ISC | 0.064323 to 0.492851 g $^{240}Pu_{eff}$ | -0.12 | -0.21 |
| Full reference range ENMC | 0.064323 to 0.492851 g $^{240}Pu_{eff}$ | -0.16 | -0.48 |

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