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**ABSTRACT**

Calorimetric assay has the reputation of providing the highest precision and accuracy of all non-destructive assay measurements. Unfortunately, non-destructive assay practitioners and measurement consumers often extend, inappropriately, the high precision and accuracy of calorimetric assay to very low mass items. One purpose of this document is to present more realistic expectations for the random uncertainties associated with calorimetric assay for weapons' grade plutonium items with masses of 200 grams or less.

# Calorimetry of Low Mass $^{239}$Pu Items

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## Introduction

Calorimetry is a non-destructive assay (NDA) technique for determining the power output of heat-producing nuclear materials. Heat-flow calorimeters measure the power output of nuclear materials based on the heat generated by decay of radioactive isotopes within the sample. A heat flow calorimeter works by sensing the temperature differential across a thermal resistance between the measurement chamber and a precisely controlled, constant temperature heat sink.

Calorimetric assay, as opposed to calorimetry, is defined as the combination of the calorimeter measurement of the power produced by a sample and the watts-to-grams conversion factor obtained from measurement of the isotopic distribution of the plutonium and other heat producing isotopes (significantly $^{241}$Am) in the sample. The calorimeter heat measurement is converted to mass using the effective specific power ($P_{eff}$, watts/gram) of the item to calculate a mass of special nuclear material (SNM). The combination of the two measurements yields a result for the total elemental plutonium mass in the sample. Note also that the overall random uncertainty for calorimetric assay must be the combined uncertainties of the power (watts) and effective specific power (watts/gram).

Calorimetric assay of plutonium-bearing samples often provides the highest precision and accuracy of all NDA techniques. The thermal power measurement is traceable to the US or other National Measurement Systems through electrical standards used to calibrate directly the calorimeters or to calibrate secondary $^{238}$Pu heat standards. Unlike other NDA techniques, calorimetry does not require physical calibration standards representative of the materials being assayed. Because the heat measurement result is independent of material, matrix type and packaging, calorimeters are well suited for nearly all plutonium samples. The downside of calorimetric assay is the length of time required for the power (calorimeter) measurement and the restrictive size of many calorimeters. Calorimetry measurements can take eight hours or longer and are dependent on the calorimeter, the material matrix, and the precision desired. By contrast, gamma ray isotopic measurements take approximately one hour.

Because of the insensitivity to material and matrix type, calorimetric assay is often used as a "referee" measurement for NDA methods that are more matrix dependent, for example, segmented gamma scanning and neutron counting. The lengthy assay times and size limitations prohibit the routine use of calorimetric assay for waste and many residue materials. However, when no other assay method is possible or when questions arise concerning the validity of a quicker measurement method, calorimetric assay is used frequently for determination of low masses of special nuclear material. An understanding of the random uncertainties for low mass items will be helpful to interpret calorimetric assay as well as data from other measurement methods.

## Experimental

Questions about the precision and accuracy for low mass SNM items and calorimetric assay first arose in the 1990's. To help resolve some of the questions and to gather actual data, a series of low wattage Pu238 heat source standards were commissioned, built and measured.

## Working Standards

A set of low wattage Pu238 heat source working standards was built from heat source grade Pu238 oxide materials. The grams of Pu238, power, and weapons grade equivalent Pu239 masses for the Pu238 standards are presented in Table 1, below.

**Table 1. Working standards**

| Standard ID | Mass Pu238 (grams) | Power (mWatts) | Power Date | Approximate Pu239 Equivalent Mass (grams) |
|---|---|---|---|---|
| NMT4-1/4A | 0.0579 | 27.3 | 29-Aug-1990 | 10.5 |
| NMT4-1/2D | 0.115 | 54.2 | 29-Aug-1990 | 20.8 |
| NMT4-1D | 0.227 | 107.3 | 29-Aug-1990 | 41.3 |
| NMT4-1/4B | 0.529 | 249.9 | 27-Sep-1990 | 96.1 |
| NMT4-1/2E | 1.060 | 500.5 | 27-Sep-1990 | 192.5 |

The approximate Pu239 equivalent mass was calculated by using the effective power for typical weapons grade material. The $P_{eff}$ value used was 2.6 mWatts/gram. This value was obtained from the standard CALEX1 and determined to be typical of weapons material.

## Measurements

The five low wattage working standards were measured in the Los Alamos Plutonium Facility Non-destructive Assay Laboratory in the production calorimeters. Five calorimeters were used for these measurements, three calorimeters (K1, K3, & K5) with five-inch diameter measurement cells and two calorimeters (K2 & K4) with seven-inch cells. In addition, several measurements were obtained with robotic loading and unloading of the calorimeters (ROBOCAL). Because the purpose of this document is provide an overall estimation of calorimeter uncertainties for low mass items, no attempt will be made to break out the data as a function of calorimeter or automation. All measurements were corrected for the Pu238 decay of the standards.

## Results

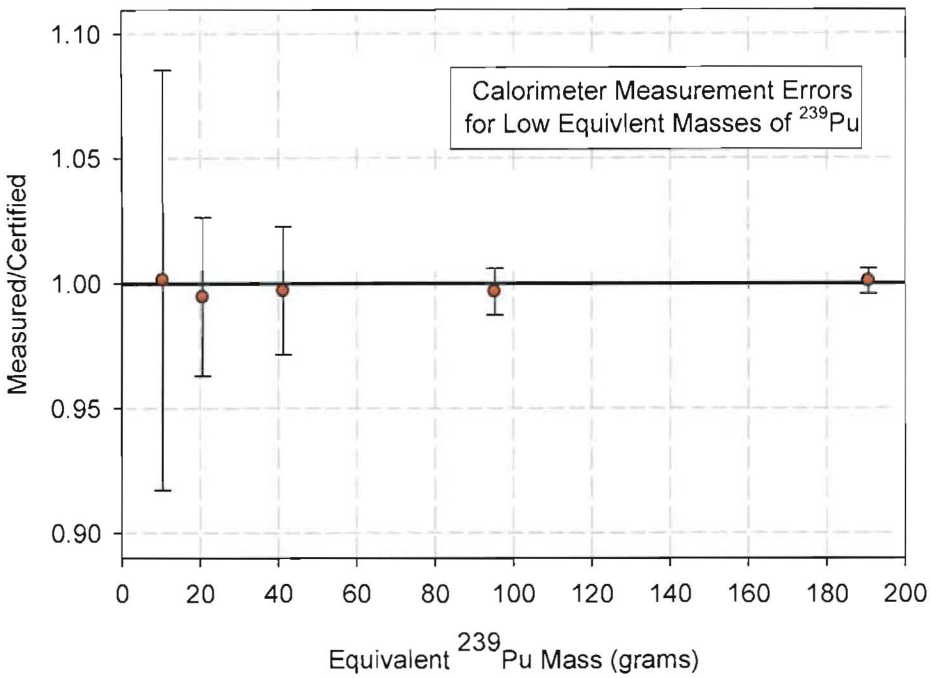
The results for the calorimetry measurements of the low wattage Pu238 samples are presented in Table 2. The single measurement standard deviation is calculated from the observed spread of the multiple measurements. The uncertainty of the standard power is 10-100 times smaller than the standard deviation of a single measurement and is neglected in the table.

**Table 2. Measurement results for low wattage standards**

| Standard | Number of Measurements | Standard Power (mWatts) | Approximate Equivalent Pu239 (grams) | Average Measured/Standard Watts | Measured/Standard Single Measurement Std Dev |
|---|---|---|---|---|---|
| **1/4A** | 29 | 27.079 | 10.42 | 1.001 | 0.0841 |
| **1/2D** | 23 | 53.77 | 20.68 | 0.995 | 0.0317 |
| **1D** | 11 | 107.3 | 41.27 | 0.997 | 0.0256 |
| **1/4B** | 22 | 247.72 | 95.28 | 0.997 | 0.00934 |
| **1/2E** | 19 | 495.59 | 190.61 | 1.001 | 0.00514 |

Results are presented graphically in Figure 1.

**Figure 1. Measurement results for low wattage working standards**




**Discussion**

The observed random uncertainties for the calorimeter power measurement decrease rapidly as the mass of the items increase. The random uncertainty (single measurement Std. Dev. in Table 2) for the 10.4 gram equivalent item is about 8.4%, 3.2% for 21 gram equivalent item, and drops to approximately 0.5% for the 200 gram equivalent item.

The random uncertainty in the power measurement does not quite translate directly to the uncertainty in calorimetric assay. As mentioned in the introduction, the calorimeter heat measurement is converted to mass using the effective specific power of the item to calculate a mass of special nuclear material (SNM). The combination of the two measurements yields a result for the total elemental plutonium mass in the sample and thus the overall random uncertainty for calorimetric assay is the combined uncertainties of the power (watts) and effective specific power (watts/gram).

It is difficult if not impossible to make many generalizations about the uncertainties in effective specific power measurements. $P_{eff}$ is calculated from the isotopic distributions and the isotopic distributions are in turn generated from gamma ray spectra. Therefore, the uncertainty in $P_{eff}$ is highly dependent on the quality of the gamma ray spectrum from which it was determined. Many factors affect the quality of spectra, including intensity of the signal, scattering in the material matrix, detector effects, and room background radiation. To estimate the $P_{eff}$ random uncertainty, the measurement history database was consulted and the $P_{eff}$ uncertainty was estimated from the $P_{eff}$ standard deviation calculated by the gamma isotopic software. All items used in the estimate were low mass items in the 10 – 200 gram range, roughly the same range as examined in this document. A random uncertainty of 0.7% was observed to be a good estimate for $P_{eff}$ for this range of materials. The uncertainty in the calorimeter measurement can be combined by standard statistical techniques with the 0.7% random uncertainty from $P_{eff}$

measurements to estimate the overall random uncertainty for calorimetric assay. The estimates are presented in Table 3.

**Table 3. Overall random uncertainty estimates**

| Equivalent Pu239 (grams) | 1 RSD Calorimeter Power Measurement (percent) | 1 RSD Gamma Isotopics $P_{eff}$ Measurement (percent) | Estimated Combined 1 RSD Uncertainty (percent) |
|---|---|---|---|
| 10.42 | 8.41 | 0.7 | 8.4 |
| 20.68 | 3.17 | 0.7 | 3.3 |
| 41.27 | 2.56 | 0.7 | 2.7 |
| 95.28 | 0.934 | 0.7 | 1.2 |
| 190.61 | 0.514 | 0.7 | 0.9 |

Note that the contribution of the $P_{eff}$ random uncertainty is not significant for Pu239 equivalent masses less than 100 grams. Conversely, as the SNM mass increases, the random uncertainty from the watts measurement (calorimeter) decreases and finally becomes a minor contribution for large mass SNM items.

Examination of the measurement history database revealed 32 items with Pu239 masses ranging from 10 – 209 grams with two or more calorimetric assay measurements. Standard deviations were computed for each of these items and the relative random variance and overall standard deviation was calculated to be 0.7%. This is consistent, in as far as there is a limited amount of relevant data in the measurement history database, with the figures in Table 3.

## Conclusions

Calorimetric assay is often used as a referee technique for segmented gamma scanning and neutron counting. However, at low masses of Pu239, 50 grams or less, the random uncertainty of the measurements can be quite large. Caution must be used for calorimetry of low mass items.