Conf-9203123--1

LA-UR- 92-883

Title: PRECISION OF GAMMA-RAY MEASUREMENTS OF THE EFFECTIVE SPECIFIC POWER AND EFFECTIVE ^{240}Pu FRACTION OF PLUTONIUM

LA-UR--92-883

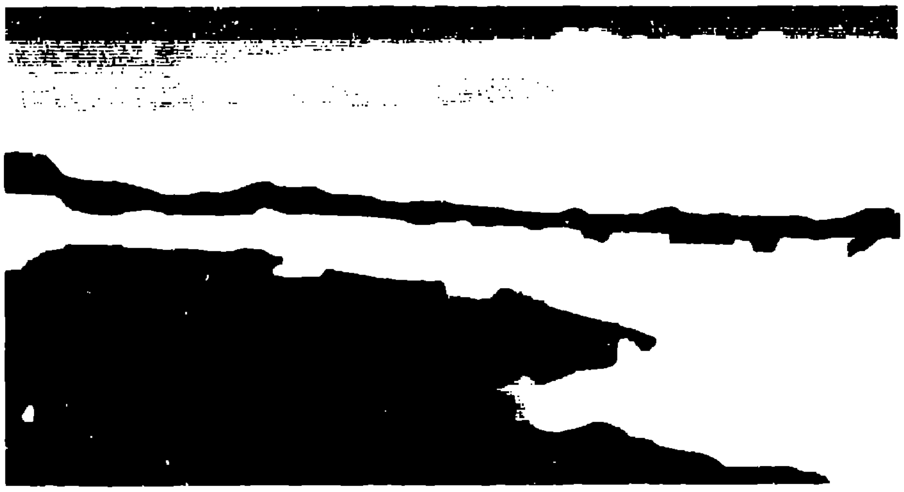
DE92 011269

Author(s): Thomas E. Sampson

Submitted to: International Workshop on Calorimetry
Ispra, Italy
March 23-27, 1992
(FULL PAPER)

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.



MASTER

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory, an affirmative action/equal opportunity employer, is operated by the University of California for the U.S. Department of Energy under contract W-7405-ENG-36. By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes. The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

PRECISION OF GAMMA-RAY MEASUREMENTS OF THE EFFECTIVE SPECIFIC POWER AND EFFECTIVE ^{240}Pu FRACTION OF PLUTONIUM*

Thomas E. Sampson
Safeguards Assay Group N-1, MS-E540
Los Alamos National Laboratory
Los Alamos, NM 87545 USA

ABSTRACT

This paper uses gamma-ray spectrometry data from replicate measurements on 40 plutonium-bearing samples to examine the repeatability of the effective ^{240}Pu fraction (^{240}Pu$_{eff}$) and the effective specific power (P_{eff}) calculated from the isotopic distribution analyzed with gamma-ray spectrometry codes. The measurements were used to identify the error component arising from repeatability in the determination of the isotopic composition of plutonium in the sample and the contribution of the error component to the uncertainty in total plutonium mass measurements from neutron coincidence counting (^{240}Pu$_{eff}$) and calorimetry (P_{eff}). The 40 samples had ^{240}Pu$_{eff}$ percentages ranging from 2 to 39% and P_{eff} values ranging from 2 to 16 mW/g Pu. Four different gamma-ray spectrometry codes (FRAM, MGA, Blue Box, and PUJRC) were used to analyze the data (not all samples were analyzed with each code).

All analyses showed that the % relative standard deviation of P_{eff} was smaller than that of ^{240}Pu$_{eff}$. This result coupled with a cursory examination of uncertainties in coincidence counting of well-characterized samples and water-bath calorimetry errors for the same types of samples leads to the conclusion that smaller uncertainties will be present in the total plutonium mass determined by the combination of calorimetry/ gamma-ray spectrometry than in the mass determined by coincidence counting/gamma-ray spectrometry. An additional examination of the biases arising from the ^{242}Pu correlation used in the gamma-ray spectrometry codes also supported this conclusion.

---

*This work is supported by the US Department of Energy, Office of Safeguards and Security.

I. INTRODUCTION

Two methods used worldwide for the nondestructive assay of plutonium are neutron coincidence counting and calorimetric assay. One must know the isotopic composition of the plutonium sample to fully interpret either measurement in units of absolute plutonium mass. Neutron coincidence counting uses the isotopic distribution to calculate the effective ^{240}Pu fraction or $^{240}\mathrm{Pu}_{\mathrm{eff}}$, which is divided into the coincidence measurement result of grams $^{240}\mathrm{Pu}_{\mathrm{eff}}$ to yield grams plutonium as

$$\text{Grams Pu} = \frac{\text{grams } ^{240}\mathrm{Pu}_{\mathrm{eff}}(\text{coincidence count})}{\text{fraction } ^{240}\mathrm{Pu}_{\mathrm{eff}}(\text{isotopic distribution})} \; . \qquad (1)$$

(The use of isotopic distributions in the calculation of multiplication corrections for coincidence counting will not be discussed here.) Calorimetry uses the isotopic distribution in a similar fashion to interpret the total wattage of a sample measured in a calorimeter by

$$\text{Grams Pu} = \frac{\text{watts (calorimetry)}}{\text{watts / g Pu (isotopic distribution)}} \qquad (2)$$

where watts/g Pu is the effective specific power, usually designated P_{eff}. A definition of $^{240}\mathrm{Pu}_{\mathrm{eff}}$ may be found in Ref. 1 while P_{eff} is defined in Ref. 2. In the remainder of this paper the term "$^{240}\mathrm{Pu}_{\mathrm{eff}}$" refers to the fraction (or percentage) of $^{240}\mathrm{Pu}_{\mathrm{eff}}$ in the sample.

The isotopic distribution measurements may be obtained from mass spectrometry on a sample* from the bulk item or, as is common in the field of nondestructive assay, from gamma-ray spectrometry measurements on the bulk, plutonium-bearing item itself. A single isotopic distribution measurement can supply the input for both P_{eff} and $^{240}\mathrm{Pu}_{\mathrm{eff}}$.

The measurement of the isotopic composition of plutonium by gamma-ray spectrometry is a well-developed technique. A review of this method may be found in Refs. 1 and 3. Reference 3, in spite of its earlier date, is somewhat more up to date than Ref. 1. Other descriptions of plutonium isotopic composition measurements and analysis codes may be found in Refs. 4-9.

Facilities using nondestructive assay techniques for assay of bulk plutonium often have the opportunity to choose between the two techniques of coincidence counting and calorimetry. Most US facilities use both but there are times when it is desirable to be able to designate the better performing technique for a given type of sample. There are as many ways of defining "better performance" as there are samples; however, we will concentrate on definitions that characterize the overall uncertainty in the final measurement result (grams of plutonium).

---

*In this paper the words sample and item are used interchangeably; this is common nondestructive assay terminology. The use of the word "sample" here is in its chemical assay sense, contrary to usage throughout the remainder of this paper.

The structure of the two expressions in Eqs. 1 and 2 shows that the final result is the quotient of two independent measurements for both coincidence counting and calorimetry. The isotopic distribution measurement is common to both methods. Each measurement result may be characterized by a systematic error and a random error. Both types of errors may be sample dependent. This last fact makes it extremely difficult to characterize the uncertainty of either method for an arbitrary sample. A recent paper[10] has tackled this ambitious task for applications important to IAEA safeguards.

This paper will illustrate, in some detail, just one of the error components in these measurement methods—the random error contributed by the gamma-ray spectrometry measurement of P_{eff} and $^{240}Pu_{eff}$. While I will also briefly discuss some of the other error components and make some comments on the analysis in Ref. 10, the main thrust will be to see how the precision (sometimes referred to as repeatability) of gamma-ray isotopic measurements of P_{eff} and $^{240}Pu_{eff}$ contributes to the uncertainty in total plutonium mass. In doing this I will examine data from replicate measurements on a wide variety of samples analyzed with several different plutonium isotopic analysis codes. I will look for trends in the magnitude of the precision of P_{eff} and $^{240}Pu_{eff}$ as a function of burnup, sample size, and analysis method and to see if any blanket statements can be made regarding the measured precision of these two quantities.

I will examine data from samples with ^{240}Pu percentages ranging from 2 to 26% and specific powers (P_{eff}) from 2.3 to over 15 mW/gPu. The analysis includes results from four different codes that analyze gamma-ray spectra to determine the isotopic composition of plutonium in a sample: FRAM[4] used at Los Alamos; MGA[5,6] used worldwide, especially in safeguards inspections; the "Blue Box,"[7,8] an older code for inspection use, now largely replaced by MGA; and PUJRC,[9] a code developed at Ispra. Not all measurements are analyzed by all codes and, indeed, it is not the purpose of this paper to compare the codes under identical conditions. What I do wish to illustrate, for any given sample analyzed with any code, is how the magnitudes of the precisions of P_{eff} and $^{240}Pu_{eff}$ compare.

II. A PREVIOUS STUDY

Some previous work in this area, focused on the broader issue of the total uncertainty in the measured plutonium mass, has been presented in Ref. 10. This study concluded that the measurement errors in the total plutonium mass were approximately the same for the combinations of coincidence counting/gamma-ray isotopics and calorimetry/gamma-ray isotopics. This result was somewhat unexpected by workers in the areas of calorimetry and gamma-ray spectrometry and prompted closer scrutiny of the data analysis methods used to obtain these results.

Several problems, mainly with assumptions in the analysis, surfaced. First, the analysis did not rigorously treat all of the error components. Only the gamma-ray spectrometry measurements were rigorously analyzed. Coincidence-counter-calibration uncertainties and sample-dependent effects were not treated. One of the two sample sets studied reported no measured or declared values for ^{241}Am, an unlikely situation considering the sensitivity of the MGA isotopics software for measuring ^{241}Am. This would tend to reduce the random errors for calorimetry/isotopics while not affecting coincidence counting. The measurement conditions for the two sets of gamma-ray spectrometry data used in the evaluation were not known to the authors of Ref. 10 and the method of treating the ^{242}Pu fraction from the gamma-ray measurements was not stated.

One of the ground rules of the study was the assignment of "operator-declared" mass spectrometry values as being without error. This meant that all differences between the gamma-ray measurements and the operator-declared values were assigned as uncertainties in the gamma-ray measurements.

Closer examination of the data revealed that the between-sample deviations for ^{238}Pu and ^{241}Am were significantly larger than the within-sample deviations. This led to the conclusion that the assumption of operator-declared mass spectrometry data as being without error was not valid. Errors in the specification of these two isotopes will affect calorimetry/gamma-ray isotopics more than they will affect coincidence counting/gamma-ray isotopics.

These problems resulted in questioning the conclusions (not the analysis methods) of the study and prompted the documentation of some of the error components for the gamma-ray spectrometry contribution to the total uncertainty in the plutonium mass obtained from these two techniques. The study in Ref. 10 should be redone without the "errorless mass spectrometry" assumption to see if the conclusions change.

First, though, it will be appropriate to note the magnitude of typical uncertainties that arise in the practice of coincidence counting and calorimetry.

III. COINCIDENCE COUNTING AND CALORIMETRY UNCERTAINTIES

Coincidence Counting. Ensslin et. al.,[11] have discussed coincidence counting uncertainties for a very wide range of sample types. Sample-dependent effects are most important for coincidence counting and can affect uncertainties by as much as a factor of 10. For well-characterized samples of metal or oxide, Ensslin quotes target measurement uncertainties in the range of 1% to 3%. He considers this to be a systematic error and notes that measurement precision (random error) for these coincidence measurements would be in the 0.3%-0.5% range [1 relative standard deviation (RSD)]. All the samples studied in this report fall into the general category of "well characterized." Measurements on other sample types are typically assigned larger uncertainties.

These values apply only to the uncertainty of the numerator in Eq. 1. They do not include any uncertainty in the determination of the plutonium isotopic composition.

Calorimetry. J. Wetzel of EG&G Mound Applied Technologies supplied this author with data from four recently delivered water-bath calorimeters. The operating range of these calorimeters varied from 0.1 to 1 watt at the low end up to 4 to 15 watts at the high end. Typical measurement precision or repeatability at the low end was about 0.5% (1 RSD) while the high-end precision was 0.05% to 0.20%. Measurement bias or systematic error from calibration is typically less than the precision. Note that these are developer's laboratory measurements. An idea of current facility practice in the US can be obtained from the annual reports of the US DOE Calorimetry Exchange Program. Two such reports, for 1988[12] and 1990[13] show nearly identical practice for measurements on the 1-watt calorimetry exchange standard. For six different laboratories, measurement precision averaged about 0.25% within a range of 0.12-0.50%. Bias was less than the precision in all cases. These numbers characterize the low end of the range of calorimetry fairly well. Measurement precision (in a relative sense) improves with increased wattage. Kilogram or larger samples of high-burnup plutonium can easily produce several tens of watts of power.

Calorimeters have the advantage of being insensitive to effects that depend on the sample composition. Generally it is considered that sample-dependent effects will not produce biases but may increase the measurement time.

This limited discussion indicates that it may be appropriate to assign approximately equal uncertainties from bias and precision, both on the order of 0.2% (relative) to water-bath calorimetry measurements (the numerator of Eq. 2). This is, of course, a simplification because calorimetry precision is a function of sample wattage.

IV. PRECISION OF GAMMA-RAY SPECTROMETRY

This section presents several examples of the precision of P_{eff} and $^{240}Pu_{eff}$ obtained from replicate measurements on a wide variety of samples and analyzed with several different isotopic analysis codes.

The first example in Table I is from measurements[14] on the CBNM NRM 271 reference material taken by this author and analyzed with both the FRAM and MGA codes. The listed standard deviations (% RSD) are computed from 20 replicates. All samples contained about 6 g of plutonium, not enough for calorimetry, but easily measured in a coincidence counter and useful to start off the comparisons. The two data sets for each code and sample represent acquisitions with different, realistic experimental conditions. For those unfamiliar with these standards, the number in the ID represents the approximate ^{239}Pu percentage.

All examples in Table I indicate that the precision of P_{eff} is better than that of $^{240}Pu_{eff}$ for these samples, measurements, and analysis codes.

The next example shown in Table II is an analysis by the FRAM code on three PuO_2 measurement control standards used routinely at Los Alamos. The number of replicates was 20 for each sample. These larger samples are routinely used to control both calorimeter and

TABLE I. CBNM Standards, FRAM, and MGA

ID	Analysis Code	P_{eff} (mW/g Pu)	$^{240}Pu_{eff}$ (%)	% RSD	
				P_{eff}	$^{240}Pu_{eff}$
Pu93	FRAM	2.5	6.4	0.22	1.56
				0.42	2.37
	MGA			0.16	0.63
				0.10	0.41
Pu84	FRAM	3.6	15.1	0.26	0.90
				0.30	1.44
	MGA			0.17	0.37
				0.13	0.29
Pu70	FRAM	10.2	23.8	0.23	1.10
				0.35	1.32
	MGA			0.43	0.62
				0.38	0.56
Pu61	FRAM	13.1	35.5	0.22	0.95
				0.31	0.99
	MGA			0.59	0.73
				0.31	0.37

TABLE II. Los Alamos Working Standards

ID	Pu mass (g)	P_{eff} (mW/g Pu)	$^{240}Pu_{eff}$ (%)	% RSD P_{eff}	% RSD $^{240}Pu_{eff}$
CALEX	400	2.5	6.0	0.30	2.05
STD40	875	3.5	12.2	0.33	1.71
LAO225BS	875	3.9	17.2	0.23	0.94

gamma-ray isotopic measurements. To put the absolute values on a meaningful basis, CALEX was measured for 1 hour at a count rate of 24 kHz while STD40 and LAO225BS were measured for 30 minutes at 40 kHz. All measurements were done with a 1-μs shaping time.

All P_{eff} precisions are less than the respective $^{240}Pu_{eff}$ precisions. The last sample in the table (LAO225BS) was also measured (20 replicates) on the Lawrence Livermore Actinide Analysis System,[15] a two-detector MGA analysis system. The % RSD for P_{eff} was 0.22 while that for $^{240}Pu_{eff}$ was 0.47.

Reference 4 also reports measurement-precision data for a wide variety of samples. These analyses with the Los Alamos FRAM code are listed for 24 different samples (including different measurements and measurement conditions for the same three samples listed in Table II). Eleven of the 24 samples had plutonium masses exceeding 60 g, large enough for calorimetry in most cases. Replicate measurements on each sample varied from 6 to 20. The observed precision for P_{eff} ranged from 0.08% to 0.33% (1 RSD) while the range for $^{240}Pu_{eff}$ was 0.68% to 3.68%. All samples gave a P_{eff} precision that was smaller than the $^{240}Pu_{eff}$ precision, as is evident from the nonoverlap of the ranges cited.

The last set of data analyzed for the precision of P_{eff} and $^{240}Pu_{eff}$ was taken during an exercise at the PERLA facility at Ispra.[9] These data consisted of a single set of replicate acquisitions on a variety of samples with ^{240}Pu contents exceeding 20%. The data were analyzed by three different isotopic codes—MGA,[5,6] the "Blue Box,"[7,8] and PUJRC.[9] Measurement results from the larger samples for the precision of P_{eff} and $^{240}Pu_{eff}$ are shown in Table III. These data had been analyzed for $^{240}Pu_{eff}$ precision and reported in Ref. 9. Table III makes the comparison with P_{eff} precision for the first time. These data add results for higher burnup plutonium that were not available in the previously discussed results. Regarding the comparison of P_{eff} precision and $^{240}Pu_{eff}$ precision—in all cases, as before, the P_{eff} precision is smaller (better) than the $^{240}Pu_{eff}$ precision, usually by a factor of two or more.

In addition to the above analysis, we also looked at the data used in the analysis presented in Ref. 10 and briefly discussed in section II above. This data consisted of replicate measurements on samples from two European facilities, Alkem and Sellafield. Sixteen samples were measured at Alkem. Only one sample had more than 6 replicates and 9 of the 16 had only 3 or 4 replicates. Twelve samples were measured at Sellafield with from 3 to 14 replicates each. Half of the samples had four or fewer replicates. The small number of replicates yielded precision results that themselves had large uncertainties. Note that the RSD of the sigma found from the distribution of n replicate measurements is

TABLE III. PERLA Standards Results

Sample ID	Pu Mass (g)	No. of Replicates	Analysis Code	P_{eff} (mW/g)	$^{240}Pu_{eff}$ %	% RSD	
						P_{eff}	$^{240}Pu_{eff}$
102	50	16	MGA	5.8	26.7	0.36	0.57
111	1000	15	MGA	5.8	26.8	0.35	0.49
		11	BLBX	5.8	27.5	0.67	1.32
		11	PUJRC	5.8	27.3	1.24	3.48
118	2500	15	MGA	5.8	26.8	0.28	0.44
		9	BLBX	5.8	27.4	0.85	1.66
		8	PUJRC	5.7	27.6	1.03	1.8
105	100	15	MGA	14.1	34.1	0.83	1.11
112	1000	15	MGA	14.1	34.2	0.73	1.71
		13	BLBX	13.9	35.4	1.53	6.34
		15	PUJRC	13.9	35.0	2.17	8.33
119	2500	15	MGA	14.1	34.2	1.13	2.07
		15	BLBX	13.9	35.1	1.38	7.67
		13	PUJRC	14.1	34.4	2.56	8.83
1007	1000	15	MGA	15.6	37.1	0.74	1.71
		11	BLBX	15.4	38.2	2.17	6.58
		15	PUJRC	15.7	36.6	3.25	7.75
120	2500	15	MGA	15.5	37.4	0.98	1.8
		16	BLBX	15.3	38.8	1.59	5.37
		15	PUJRC	15.4	38.9	3.55	7.18
CBNM70	6	16	MGA	9.9	23.4	0.49	0.82

$$\sigma_{rel}(sigma) = \frac{1}{\sqrt{2 \bullet (n-1)}} \ .$$

The Sellafield data exhibited an average precision for all 12 samples of 0.81% (1 RSD) for P_{eff} while that for $^{240}Pu_{eff}$ was 1.25%. In contrast to all of the previously described measurements, 2 of the 12 samples showed individual average results in which the % RSD for P_{eff} was larger than % RSD for $^{240}Pu_{eff}$. Results from the 16 Alkem samples were similar to those from the Sellafield data: the average P_{eff} precision was 0.89% while that for $^{240}Pu_{eff}$ was 1.34%. For 6 of the 16 Alkem samples the % RSD for P_{eff} was larger than the % RSD for $^{240}Pu_{eff}$. We performed a simple sign test on these results, testing the hypothesis that the ^{240}Pu$_{eff}$ precision equaled the P$_{eff}$ precision. The Sellafield data set taken by itself and the two sets of data combined yielded a statistically significant result indicating ^{240}Pu$_{eff}$ % RSD was greater than the P$_{eff}$ % RSD. This test, for the Alkem data alone, did not indicate statistical significance. Some additional statistical tests that also consider the varying small number of replicates may be warranted here.

V. EFFECT OF ^{242}Pu BIAS

Gamma-ray spectrometry cannot directly measure the ^{242}Pu abundance because of the isotope's long half-life and the low abundance of the gamma rays from this isotope. Instead, a correlation is usually used to relate the ^{242}Pu abundance to the abundances of the other directly measured isotopes.[16] Correlations can be a function of the reactor history as well as the reactor type and no universal expression has been found that is best for all samples. Because of this, large biases (10%–50%) can be present in the ^{242}Pu abundances reported by gamma-ray spectrometry measurements. These biases will, in turn, contribute to bias in both P$_{eff}$ and ^{240}Pu$_{eff}$. While ^{242}Pu does not directly influence P$_{eff}$, it affects its magnitude through the normalization condition that all plutonium isotopic abundances must sum to 100%. Thus a bias in ^{242}Pu propagates into a bias in all the other isotopes which, in turn, causes a bias in P$_{eff}$. The situation for ^{240}Pu$_{eff}$ is somewhat different in that ^{242}Pu appears directly in the expression for ^{240}Pu$_{eff}$ as well as affecting the normalization.

Figure 1 below shows the result of a sensitivity study using isotopic distributions from a subset of the samples discussed in this paper. For this study the correct isotopic distribution was used to calculate both P$_{eff}$ and ^{240}Pu$_{eff}$. Then the ^{242}Pu abundance was increased by 10%, the isotopic distribution was renormalized, and P$_{eff}$ and ^{240}Pu$_{eff}$ were recalculated. The fractional difference in P$_{eff}$ and ^{240}Pu$_{eff}$ between the two cases is plotted against the magnitude of ^{240}Pu$_{eff}$ in Fig. 1. The result is that ^{240}Pu$_{eff}$ is more sensitive to a bias in ^{242}Pu abundance than is P$_{eff}$.

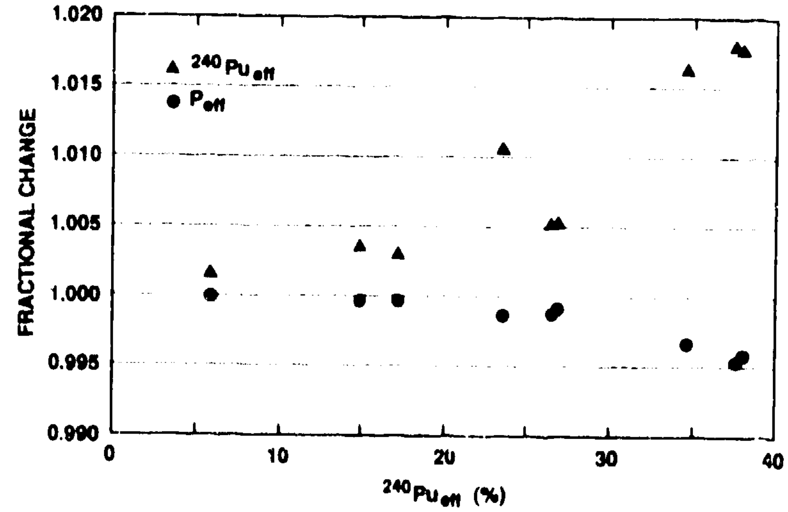


Fig. 1. Fractional change in ^{240}Pu$_{eff}$ and P$_{eff}$ for a +10% change in ^{242}Pu.

Some inspection scenarios permit the inspector to use the operator's declared value for ^{242}Pu; this removes ^{242}Pu as a source of bias from both techniques. All comparisons in this paper used mass spectrometry values for ^{242}Pu. The ^{242}Pu bias effect discussed in this section was not present in the analysis for P_{eff} and $^{240}Pu_{eff}$ presented in section IV. In any case, if such bias were present, it would not affect the *precision* calculated from replicate measurements.

VI. SUMMARY AND CONCLUSIONS

I have examined several error components that contribute to the total uncertainty of the plutonium mass as determined by the combinations of coincidence counting/gamma-ray spectrometry and calorimetry/gamma-ray spectrometry. I have presented numerous examples, which illustrate that the precision of P_{eff} from gamma-ray spectrometry is better (smaller % RSD) than that of $^{240}Pu_{eff}$ from the same measurement. I have also briefly discussed some examples from the literature that illustrate some of the typical uncertainties arising in calorimetry and neutron coincidence counting. While this discussion was not rigorous or exhaustive, it strongly indicated that water-bath calorimetry uncertainties (both bias and random errors) are likely to be smaller than the corresponding uncertainties for coincidence counting on well-characterized samples of appropriate size for calorimetry.

An examination of a recently published analysis showing roughly equal total plutonium uncertainties for coincidence counting and calorimetry was found to be flawed in several areas creating reasonable doubt about the validity of the study's conclusions.

The sensitivity of both P_{eff} and $^{240}Pu_{eff}$ to biases in ^{242}Pu, often present in gamma-ray spectrometry measurements, was also examined. Data from a representative set of samples showed that $^{240}Pu_{eff}$ is affected more than P_{eff} by ^{242}Pu bias.

Taken together, these results present persuasive evidence that the uncertainties of plutonium mass determination from water-bath calorimetry/gamma-ray spectrometry should almost always be less than those from coincidence counting/gamma-ray spectrometry for well-characterized samples. It must also be noted that considerations other than total uncertainty are often paramount when choosing a measurement technique for a particular problem. The generally shorter measurement time for coincidence counting, relative to calorimetry, is often an overwhelming consideration when choosing a technique.

It is also noted that while this study considered "well-behaved" materials, the nature of the sample greatly affects coincidence counting uncertainties—newer coincidence counting methods and analysis methods can reduce bias on "difficult" samples. The use of air-bath calorimeters may change the error estimates for calorimetry as well as reduce measurement time. Certain categories of scrap and waste samples adversely affect the bias of the gamma-ray isotopic measurements but are not seen to change the relative differences in the % RSD between P_{eff} and $^{240}Pu_{eff}$. All these conditions deserve additional study for the application to difficult, heterogeneous samples and other special cases.[17].

ACKNOWLEDGMENTS

I wish to thank Larry Bruckner for his assistance with the statistical analysis of the Sellafield and Alkem data presented at the end of section IV.

REFERENCES

1. T. D. Reilly, N. Ensslin, and H. A. Smith, Jr., Eds., *Passive Nondestructive Assay of Nuclear Materials*, NUREG/CR-5550 (U.S. Nuclear Regulatory Commission, Washington DC, 1977), p. 457.

2. "Plutonium-Bearing Solids — Calibration Techniques for Calorimetric Assay," ANSI N15.22-1987 (American National Standards Institute, Inc., New York, 1987).

3. T. E. Sampson, "Plutonium Isotopic Composition by Gamma-Ray Spectroscopy: A Review," Los Alamos National Laboratory report LA-10750-MS (September 1986).

4. Thomas E. Sampson, George W. Nelson, and Thomas A. Kelley, "*FRAM*: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse Height Spectra," Los Alamos National Laboratory report LA-11720-MS (December 1989).

5. R. Gunnink, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances. Volume 1, Methods and Algorithms," Lawrence Livermore National Laboratory report UCRL-LR-103220, Vol. 1 (ISPO No. 317, Task No. A.161) (April 1990).

6. R. Gunnink and W. D. Ruhter, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundances. Volume 2, A Guide to Using MGA," Lawrence Livermore National Laboratory report UCRL-LR-103220, Vol. 2 (ISPO No. 317, Task No. A.161) (September 1990).

7. W. D. Ruhter, "A Portable Microcomputer for the Analysis of Plutonium Gamma-Ray Spectra, Vol. I. Data Analysis Methodology and Hardware Description." Lawrence Livermore National Laboratory report UCRL-53506, Vol. 1 (ISPO-209) (May 1984).

8. W. D. Ruhter, "A Portable Microcomputer for the Analysis of Plutonium Gamma-Ray Spectra, Vol. II. Software Description and Listings," Lawrence Livermore National Laboratory report UCRL-53506, Vol. 1 (ISPO-209) (May 1984).

9. D. D'Adamo, M. Franklin, S. Guardini, C. Vicini, R. Gunnink, W. Ruhter, and G. Varasano. "Performance Evaluation of Gamma Spectrometry Codes for International Safeguards," *Nucl. Mater. Manage.* **XIX**, 782-791 (1990).

10. P. DeRidder, G. Laszlo, A. S. Goldman, "Comparison between Calorimeter and HLNC Errors," *Nucl. Mater. Manage.* **XX**, 767-771 (1991).

11. N. Ensslin, D. G. Langner, H. O. Menlove, M. C. Miller, and P. A. Russo, "Some Target Assay Uncertainties for Passive Neutron Coincidence Counting," *Nucl. Mater. Manage.* **XIX**, 610-614 (1990).

12. "Calorimetry Exchange Program Annual Data Report for 1988," EG&G Mound Applied Technologies report MLM-MU-89-68-0003.

13. "Calorimetry Exchange Program Annual Data Report for 1988," EG&G Mound Applied Technologies report MLM-MU-91-63-0005.

14. R. J. Friar and T. E. Sampson, "Plutonium Isotopic Abundance Measurements on CBNM NRM 271 Analyzed with the FRAM and MGA Codes," Los Alamos National Laboratory report to be published.

15. Zachary M. Koenig, Wayne D. Ruhter, Ray Gunnink, William M. Buckley, and David C. Camp, "Actinide Isotopic Analysis System for Los Alamos Plutonium Handling Facility," *Nucl. Mater. Manage.* **XX**, 470-473 (1991).

16. R. Gunnink, "Use of Isotope Correlation Techniques to Determine ^{242}Pu Abundance," *Inst. Nucl. Mater. Manage.* **IX** (2), 83-93 (1980).

17. H. O. Menlove, R. Abedin-Zadeh, and R. Zhu, "The Analyses of Neutron Coincidence Data to Verify Both Spontaneous-Fission and Fissionable Isotopes," Los Alamos National Laboratory report LA-11639-MS (August 1989).