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CONF-980733--

Title: **INVENTORY VERIFICATION MEASUREMENTS USING
NEUTRON MULTIPLICITY COUNTING**

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Submitted to: **1998 INMM
Naples, FL USA
July 26-30, 1998
(FULL PAPER)**

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**INVENTORY VERIFICATION MEASUREMENTS USING NEUTRON
MULTIPLICITY COUNTING**

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*Presented at the
Institute of Nuclear Materials Management
39th Annual Meeting
Naples, Florida USA
July 26-30, 1998*

Inventory Verification Measurements Using Neutron Multiplicity Counting*

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Abstract

This paper describes a series of neutron multiplicity measurements of large plutonium samples at the Los Alamos Plutonium Facility. The measurements were corrected for bias caused by neutron energy spectrum shifts and nonuniform multiplication, and are compared with calorimetry/isotopics. The results show that multiplicity counting can increase measurement throughput and yield good verification results for some inventory categories. We provide recommendations on the future application of the technique to inventory verification.

INTRODUCTION

Department of Energy (DOE) Order 5633.3B, Chapter II, Section 3d describes the inventory verification/confirmation measurement requirements for DOE facilities: "At each facility, management shall establish and implement a system for performing measurements as part of a physical inventory. Verification measurements shall be made on special nuclear material items that are not tamper-indicating. Separate sampling plans shall be implemented for verification and confirmation measurements to assure that a sufficient number of nontamper-indicating items are measured." Based on these requirements, the current goal of the inventory verification program at the Los Alamos Plutonium Facility is to ensure the integrity of the inventory at the 95% confidence level for 90% of the material in the inventory.

At the Plutonium Facility, verification measurements are performed using neutron coincidence counters, segmented gamma scanners, and calorimetry/gamma-ray isotopics. In order to minimize inventory differences introduced by instrument bias, items containing less than 200 g total plutonium are sent for neutron or gamma assay, while those items containing above 200 g are sent for calorimetry/gamma-ray isotopics.

Based on a recent agreement with the DOE Albuquerque Area Office, the Los Alamos Plutonium Facility has a yearly obligation to perform 630 total verification measurements. These measurements are comprised of a pool of randomly selected items and a nonrandom selection of poorly measured, attractiveness level C and D items. From an internal assessment of the inventory, it is projected that 230 of these verification measurements will require calorimetry/gamma-ray isotopics analysis under the current measurement strategy. When these measurements are added to the projected number of programmatic measurements required to support work at the plutonium facility, there is a shortfall in the available capacity of calorimetry/gamma-ray isotopic measurements.

Extending the measurement range for neutron assay above the 200-g cut-off using neutron multiplicity counting would help reduce the overload on the calorimetry/gamma-ray isotopic capacity. Typical calorimetric assays take from 8 to 12 h, plus one hour for the gamma-ray isotopic measurement. A typical neutron multiplicity measurement takes one-half to one hour. The use of neutron multiplicity counting as a replacement for calorimetric assays could reduce the measurement time by as much as 12 h for those items where multiplicity counting is applicable.

To determine the potential application of multiplicity counting to inventory verification, we carried out multiplicity measurements of 50 large plutonium items with good calorimetry/isotopics values. This paper describes the measurement equipment used, the calibration and data analysis procedure for the multiplicity counter, and the results obtained. Several state-of-the-art data-correction procedures were applied to the multiplicity analysis, and these are described in detail. We summarize the results obtained, and offer our recommendations on the use of multiplicity counting for this purpose.

IN-PLANT MEASUREMENTS

The neutron multiplicity measurements made for this report used the In-Plant Multiplicity Counter, also called the Pyrochemical Multiplicity Counter. This counter was designed for in-plant use, and the parameters identified as important for multiplicity assay were optimized (Ref. 1). It has 126 ^3He tubes arranged in four rings with an absolute neutron detection efficiency of 56% and a very flat energy response profile and spatial response profile (Ref. 2). The in-plant counter has previously been used in the Los Alamos and Lawrence Livermore National Laboratory Plutonium Facilities to assay inventory items (Refs. 3 and 4). The counter contains 36 Amptek preamps, and has a relatively low multiplicity dead time.

The calorimetry measurements used as reference assays for this report were made using high-precision water-bath calorimeters in operation at the Plutonium Facility. All of the calorimeters are currently certified to perform plutonium accountability measurements. At the time of the assays, the calorimeters were demonstrated to be "in-control," as defined by the Los Alamos measurement control program, through weekly measurements of certified ^{238}Pu heat standards. The precision of the heat measurement was typically 0.1% to 0.3%.

The isotopic composition of the items was determined by high-resolution gamma-ray spectroscopy, using the FRAM analysis code (Ref. 5). Sample measurement times were in the range of 3600 to 7200 s. The precision of the specific power determination, which yields total plutonium mass, ranged from 0.3% to 1.0%, with typical values near 0.6%. The precision of the ^{240}Pu -effective determination, which is the measured quantity in multiplicity counting, ranged from 1.1% to 4.6%, with typical values near 3.0%.

Tables I and II summarize the 50 measurements made in this study. Table I includes the 10 calibration standard measurements made over the course of the study for measurement control and calibration purposes, and the 13 measurements on metal items. The metal items are listed in order of increasing sample multiplication M . Table II includes measurements of 27 plutonium oxides, impure oxides, and process residues. These are listed in order of increasing α , the ratio of (α, n) to spontaneous fission neutrons.

In both Tables I and II, the first column indicates the sample identification, and the second column provides a brief description of the sample. For 47 of the measurements, the reference plutonium and reference ^{240}Pu -effective values in the third and fourth columns are those determined from calorimetry and FRAM isotopic analysis. For three of the items, reference values are based on less-accurate techniques.

NEUTRON MULTIPLICITY COUNTER CALIBRATION PROCEDURE

The calibration procedure for neutron multiplicity counting does not require a series of representative physical standards. Instead, the initial calibration can be done with only californium

neutron sources. However, the full procedure that can be implemented is actually quite complex, and is still being refined. One of the goals of this study was to continue the development of this procedure, and to document it for use at other facilities. The calibration procedure used here consisted of the following steps.

1. A series of californium sources (the Los Alamos "rainbow" series) was used to determine the dead-time corrections for singles, doubles, and triples for the multiplicity counter. This was done by adjusting the dead-time coefficients until the observed doubles/singles and triples/doubles ratios for the sources were constant, independent of source strength. To use this procedure, the curium and ^{250}Cf content of the sources must be low so that the true ratios will be constant. For this counter, the multiplicity dead-time coefficient was found to be 90 ns and the double coincidence coefficient A was 0.36 μs .
2. Using one of the californium sources of known yield, the dead-time-corrected singles, doubles, and triples count rates were used to determine initial values for the absolute neutron detection efficiency ϵ , the doubles gate fraction f_d , and the triples gate fraction f_t . These three quantities constitute the "calibration coefficients" for neutron multiplicity counting within the framework of the "point model," which assumes that neutron behavior is constant across the sample, as though it were a single point in space.
3. At this stage, one should correct for the difference in efficiency between californium and plutonium by Monte Carlo calculations or by measuring an actual plutonium standard. For this counter, all three calibration coefficients were adjusted with actual plutonium standards, as described in the next step.
4. Whenever possible, the three calibration coefficients should be adjusted for errors due to point-model assumptions by measuring one or more actual plutonium standards similar in size, shape, density, and mass to the actual unknown samples to be assayed. The results of this assay can be used in several different ways, including
 - a. Adjust f_t only, since it is most sensitive to point-model assumptions, and most sensitive to errors in nuclear data coefficients and energy spectrum shifts. The adjustment may be on the order of 10%, with the final assay changing by a similar amount.
 - b. Adjust ϵ only to allow for scattering and moderation in distributed samples.
 - c. Adjust ϵ , f_d , and f_t in some consistent way, not yet defined, with one standard. After adjustment, sample mass, M , and α should all be correct to the extent to which they are known. Otherwise, the assay of actual samples will introduce a bias that increases as M or α increases.

For this counter, the calibration coefficients were initially adjusted using the LAO and PEO series of standards to obtain $\epsilon = 0.5830$, $f_d = 0.4377$, and $f_t = 0.1945$ (Ref. 3). Later, the counter was moved several times, which may cause a change in efficiency because the counter body consists of two halves that may not fit together again in the same way. For this study, we used a series of 10 measurements of the Los Alamos Calex standard and two other standards to adjust the efficiency to $\epsilon = 0.5706$. The gate fractions were not adjusted, because there is no independent information on the M and α values of these standards that would provide a physical basis for adjustment.

5. Then, we corrected all multiplicity assays for shifts in the neutron energy spectrum due to neutrons from (α ,n) reactions by using the ratio of counts in the inner and outer rings. This bias is visible in Fig. 1 as an overall downward tilt of the observed, uncorrected assay/reference values plotted as a function of ring ratio. The ring-ratio correction is not yet installed in the neutron coincidence counter (NCC) software code, and must be applied manually. This can be done in two ways:

- a. An empirical correction can be applied based on the observed relationship between ring ratio and assay bias in Fig.1.
- b. An iterative mathematical correction can be applied which subtracts the estimated spontaneous fission contribution to the signal in the inner and outer rings, determines a ring-ratio correction from the remaining signals, and then solves the multiplicity equations with separate detection efficiencies and fission probabilities for the (α ,n) neutrons (Ref. 6).

We have used the first approach for this study.

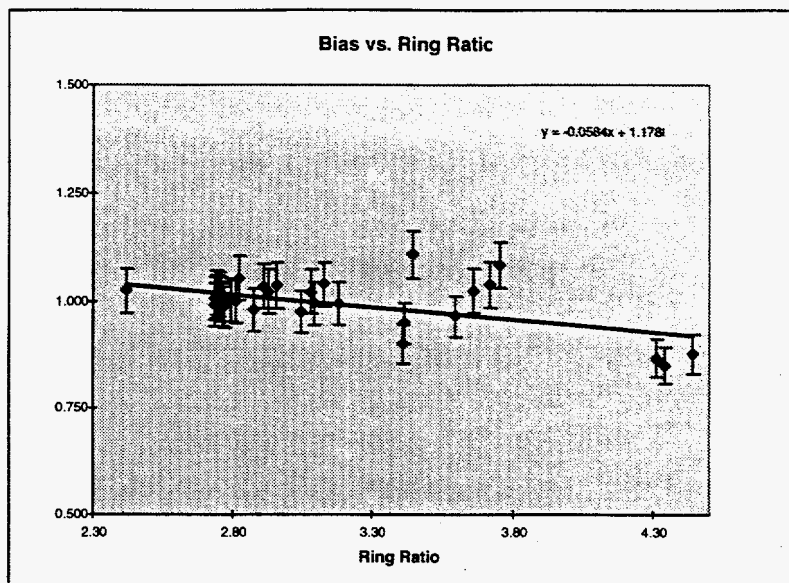
6. For large metal samples, a correction for the nonuniform probability of fission in large metal plutonium items is required. The correction can be based on the observed sample multiplication or on the ratio of quad to triple coincidences. We investigated both approaches, as described below. This correction can be applied before or after the correction for energy spectrum bias described in step 5 above. For this study, we applied the energy spectrum bias correction first to all of the measurements.

7. There is a final step that can be done at the end of a large measurement campaign if other defensible documentation such as calorimetry measurements or destructive analysis show that the multiplicity assays have a small overall residual bias. Then the calibration can be adjusted in one of the following ways:

- a. Adjust f_t only.
- b. Adjust ϵ , f_d , and f_t in some consistent way, not yet defined, to minimize the average bias.

Of necessity, this adjustment would be intended to remove an overall bias in sample mass only, not M or α . We have not applied such an adjustment, but the final results in Tables II show that a correction of several percent could be applied to the impure plutonium items to reduce the bias relative to calorimetry.

Fig. 1. Assay bias as a function of observed detector ring ratio for calibration standards, and impure items.



RESULTS OF ENERGY SPECTRUM BIAS CORRECTION

Figure 1 illustrates the observed assay bias as a function of the observed detector ring ratio for 10 calibration standards and 25 pure and impure plutonium items. This bias is due to the fact that neutrons from (α ,n) reactions may have significantly different average energies than spontaneous fission neutrons, and therefore have different detection efficiencies and fission probabilities. In the multiplicity counter, the inner ring (Ring 1) has a high detection efficiency for low-energy neutrons, and the outer ring (Ring 4) has a high detection efficiency for high-energy neutrons (Ref. 7). Thus the ratio of Ring 1/Ring 4 is a good measure of the average neutron energy in the counter and can provide a correction for this bias.

In Fig. 1, the sloped line superimposed on the data is a least-squares-fit of the bias as a function of ring ratio. This correction was applied to all of the 50 measurements made in this study, including the large metal items, and is included in Tables I and II. (The correction was normalized to give no change to the Calnex standard, which was taken as the reference for calibration.) Past experience has shown that metal items have a slightly softer energy spectrum and a higher ring ratio than oxide items (Ref. 4). This effect is relatively small, but does shift the metal assays by an average of about 2% in this study. We applied the ring-ratio correction to the metal items before applying the multiplication bias correction described below. However, we should note that in principle the measurement effects are coupled, and that neither correction is complete or exact by itself.

RESULTS OF MULTIPLICATION BIAS CORRECTION

Past measurements of large metal samples have demonstrated the need to correct multiplicity analysis based on the three-parameter point model for the nonuniform probability of fission in large metal plutonium items (Ref. 4). Monte Carlo calculations have verified this effect, and an empirical correction based on the observed sample multiplication has been developed. Figure 2 illustrates the dependence of assay bias on sample multiplication for the standards and metals measured in this study, after applying the energy bias correction using the ring ratios.

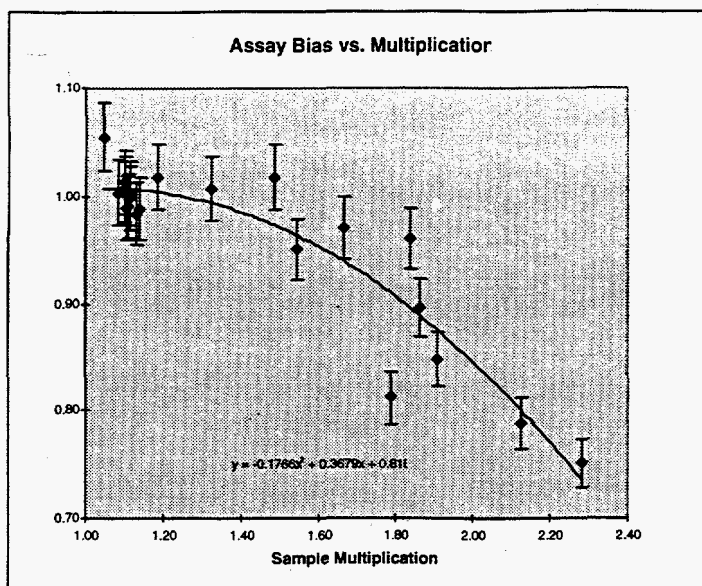


Fig. 2. Assay bias as a function of sample multiplication for standards and metals, after correction for energy bias using the observed ring ratios.

Because the quad coincidence rate is even more sensitive to sample multiplication than the triple coincidence rate, we have begun to investigate the use of the quads/triples ratio as another potential correction method (Ref. 8). To further this investigation, we analyzed the multiplicity data obtained in this study to obtain the quad coincidence count rate.

The first challenge in working with quad coincidences is to apply a dead-time correction to the data, because a formal dead-time correction for quad coincidences has not yet been developed. For this analysis, we applied an empirical correction to the quad coincidences, as summarized in Table III. The table lists the measured singles for several standards and for the metal items. Based on past experience, the doubles correction factor is 4 times the singles correction factor. The triples correction factor has a larger exponent than the doubles correction factor by the ratio $\ln(\text{TCF})/\ln(\text{DCF})$, which varies from about 2.5 for low alpha, high mass samples to 5 for very high alpha, high count rate samples. For the large metal samples of most interest here, we used the ratio 2.5. Then we assumed that the exponent for the quads correction factor would be larger than for the doubles correction factor by the ratio 4 instead of 2.5, so that the quads correction factor would be the triples correction factor raised to the 1.6 power.

Further study is required to see if this empirical quads dead-time correction is generally applicable. For this data set, the resulting quads are in good agreement with Figure-of-Merit calculations of the expected count rate (Ref. 8). The dead-time-corrected count rate is typically within 5% to 10% of the calculated count rate, even when the dead-time correction is nearly a factor of 2. The relative standard deviation (RSD) of the quads count rate is good (typically 1%) for large metal samples and is usually within 20% to 50% of the calculated RSD, depending on the magnitude of the sample multiplication, which is consistent with the behavior described in Ref. 8. Also, the quads/triples ratio behaves in a smooth way as mass and alpha changes, as illustrated below.

Figure 3 illustrates the dependence of assay bias on the quads/triples ratio for the standards and metals. The dependence is remarkably similar to that in Fig. 2, suggesting that the observed quads/triples ratio provides essentially the same information as the observed sample multiplication. This is verified in Fig. 4, which plots the quads/triples ratio vs sample multiplication. This close relationship may be useful in the future for other purposes, but for this study we again used a bias correction based on sample multiplication. Table I applies this correction to the metal items. (Other

potential correction algorithms include the use of the quads alone, or the use of the quads/doubles-squared ratio. These were plotted, but do not show a good correlation with assay bias.)

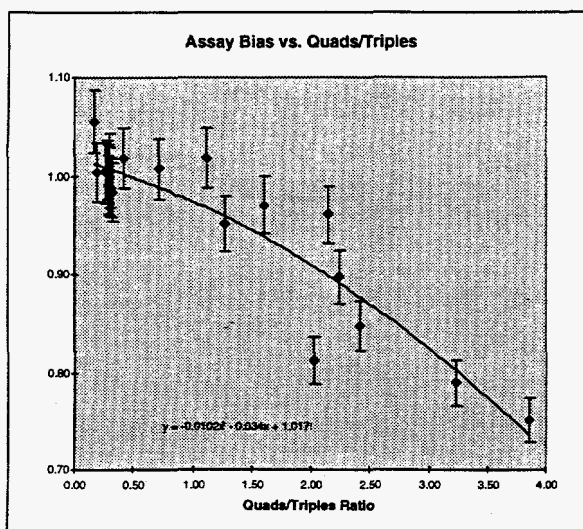


Fig. 3. Assay bias as a function of quads/triples ratio for standards and metals, after correction for energy bias using the observed ring ratios.

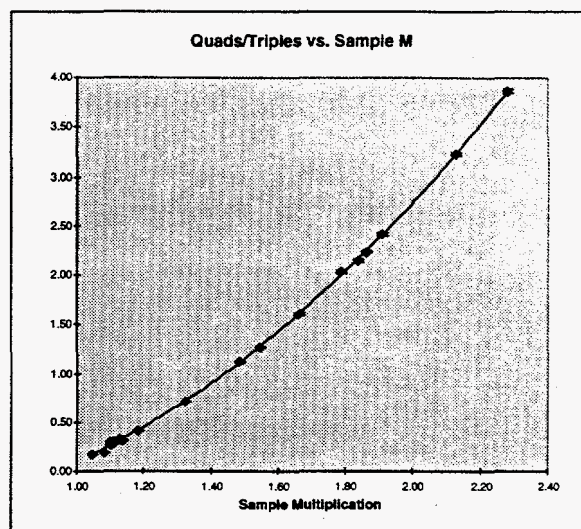


Fig. 4. Quads/triples ratio vs sample multiplication for standards and metals.

SUMMARY OF ASSAY RESULTS

Table I summarizes the observed precision and bias for a series of calibration standards measured over the course of one year. The RSD of each individual measurement is in the range of 0.5% to 1.0%, and there is no noticeable drift. The average assay bias over all 10 measurements is close to 0%, because the calibration was normalized to these standards after the energy bias correction was applied.

The effect of the energy bias and multiplication bias corrections on the metal measurements are also shown in Table I. The original scatter on the ratio of (plutonium mass by multiplicity assay)/(plutonium reference mass by calorimetry/gamma isotopics) was $-9.1\% \pm 10.2\%$ RSD. Application of the energy bias correction reduced this scatter slightly to $-7.2\% \pm 9.9\%$ RSD, and application of the multiplication bias correction reduced this scatter to $1.3\% \pm 4.6\%$ RSD. With stream average isotopics rather than gamma-ray isotopics, the final scatter is $-1.8\% \pm 3.3\%$ RSD.

Table II summarizes the multiplicity assays of the pure oxide, impure oxide, and process residue items. The table segregates the items into 18 with α values of 6 or less, and nine with α values greater than 6. This segregation level is somewhat arbitrary, but we can see that for count times of 1800 to 3600 s and α in the range of 1 to 6, the precision in the measured ^{240}Pu -effective mass is 1% to 3%, which is comparable to the uncertainty in the determination of ^{240}Pu -effective by gamma-ray isotopics. For α in the range of 7 to 34, the individual assay precision is in the range of 4% to 47%, and becomes the limiting factor in the potential assay accuracy. So for high α samples, multiplicity results are not useful in reasonable counting times, and calorimetry is the preferred option if the container sizes are appropriate.

A number of anomalous or outlier results are observed in Table II. Three items may be outliers because their reference mass values are not based on calorimetry/gamma isotopics. The reference value for RESLC-6 is based on segmented gamma scanning, the reference value for ARF876595 is

based on chemical assay, and the reference value for GVB20-M99 (the PuBe neutron source) is based on shipper declaration. In addition, item ASHRCVP83 shows a large bias, even though it was measured twice by calorimetry. On the other hand, the ring ratio, α , and multiplication values are reasonable values for multiplicity assay. Possible explanations include electronic noise in the multiplicity counter or the presence of curium in the ash. The remaining anomaly is item PEOF13, which may have a problem with its measured gamma-ray isotopic composition, which is not consistent with its expected material type. Some of the measured items were in very large containers, so that the plutonium may have been well away from the center of the assay chamber in the multiplicity counter. Although the counter was designed to have a flat efficiency profile, we believe that it is important to note unusual placements in the future.

We have excluded the above five outliers in computing the typical scatter expected in multiplicity measurements of impure plutonium items. Then, for 16 of the 18 low α measurements in Table II, the original scatter on the ratio of (plutonium mass by multiplicity assay)/(plutonium reference mass by calorimetry/gamma isotopics) was $-1.6\% \pm 5.7\%$ RSD. Application of the energy bias correction reduced this scatter slightly to $1.5\% \pm 4.5\%$ RSD. With stream average isotopics rather than gamma-ray isotopics, the final scatter is $0.4\% \pm 10.9\%$ RSD. A multiplication bias correction is not significant at these low multiplication values and was not applied.

For six of the nine high α measurements in Table II, the original scatter on the ratio of (plutonium mass by multiplicity assay)/(plutonium reference mass by calorimetry/gamma isotopics) was $-9.2\% \pm 18.8\%$ RSD. Application of the energy bias correction yielded $-4.0\% \pm 18.9\%$ RSD. With stream average isotopics rather than gamma-ray isotopics, the final scatter is $-7.5\% \pm 20.7\%$ RSD. For these high α items, the measurement precision in 1800 to 3600 s is clearly limited to about 20%. For example, item INCA-19 was measured twice. The 1800-s measurement showed a large bias, but was not inconsistent with the 47% statistical precision of the measurement. The 14340-s measurement had 10% precision, and an even smaller bias, illustrating the fact that multiplicity counting can assay high α samples if necessary, using very long counting times.

CONCLUSIONS AND RECOMMENDATIONS

This paper has described the measurement results from a diverse set of items at the Los Alamos Plutonium Facility. Based on the analysis of the data carried out so far, we offer the following conclusions and recommendations.

1. It is helpful to segregate items being considered for multiplicity counting into the categories identified in this study: calibration and measurement control standards, metal, low α plutonium, and high α plutonium ($\alpha > 6$). These categories can be defined by the observed assay results for sample multiplication, mass, α , or measurement precision.
2. For high α plutonium, multiplicity counting can be useful, but is not the preferred technique because of the long counting times required. For the other categories, counting times of 1800 s (half hour) are usually sufficient to eliminate counting statistics as a significant contribution to the overall assay precision. It may be helpful to do all assays at 1800 s, then decide on the basis of the observed α whether additional counting time is warranted.
3. The multiplicity counter requires a careful calibration procedure based on a combination of californium sources and one or more physical standards to obtain optimum results. Then it is possible to obtain good results for a wide variety of in-plant materials without representative standards.

4. For metal items, the data analysis procedure should include at least a multiplication bias correction, and for impure plutonium items the procedure should include at least an energy bias correction. It is feasible to apply both a multiplication bias and an energy bias correction to most items, but the corrections are actually coupled, and an exact, combined correction procedure has not yet been developed.
5. The overall scatter of the multiplicity assays was about 5% RSD for metals, 5% RSD for low α samples, and 20% RSD for high α samples with $\alpha > 6$. The overall assay precision of multiplicity counting for total plutonium mass has a lower limit of about 3% RSD once the error on the ^{240}Pu -effective as determined by gamma-ray isotopics is folded in. By way of comparison, the reference total plutonium masses have an overall error of about 0.5% from calorimetry and gamma isotopics. This limits the potential performance of multiplicity counting relative to calorimetry if the isotopic composition is determined from gamma-ray analysis.
6. In addition to the typical performance numbers quoted above, we can expect some fraction of the measurements (10% in this study) to be well outside the reasonable expected limit of error for multiplicity counting, either because the reference values are less accurate than multiplicity counting or because of other unresolved discrepancies.
7. The use of stream average isotopics can provide good results for most of the items measured in this study and can eliminate the time required to do gamma-ray isotopics and the error due to the determination of ^{240}Pu -effective by gamma-ray isotopics. With stream average isotopics, the scatter in the multiplicity assays was about 3% RSD for metals, 11% RSD for low α samples, and 21% RSD for high α samples.
8. Overall, if the multiplicity assays had been used for verification purposes, the result would have been that 85% of the metals and 67% of the other items would have been verified to within 5%. Using stream average isotopics, the result would have been that 92% of the metals and 56% of the other items would have been verified to within 5%.
9. Multiplicity counting can be a useful technique for inventory verification. Based on the results of this study, it should be possible to verify about 75% of a diverse inventory of plutonium metals, oxides, salts, and residues to about 5% accuracy in one-half hour counting times. This would reduce the number of items that require calorimetry and gamma-ray isotopic analysis and allow an increase in facility measurement throughput.

ACKNOWLEDGMENTS

We would like to acknowledge the NDA laboratory and vault personnel at the Los Alamos Plutonium Facility for their efforts in acquiring the calorimetry and gamma-ray isotopics data used in this study.

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

Table I. Multiplicity Results for Standards and Metals.xls

Sample ID	Sample Type	Reference Pu total (g)	RSD on spec. power	Reference Pu-240eff (g)	RSD on Pu-240 effective	Count Time (s)	Quads/Triples Ratio	Inner/Outer ratio	Sample M	Sample alpha	Pu-240 eff mass (g)	Pu-240 RSD (g)	assay/ refer. (%)	Pu-240 w/Energy Bias Corr	assay/ refer. (%)	Pu-240 w/Mult Bias Corr	assay/ refer. (%)	Strm. Avg. Pu-240eff (g)	assay/ refer. (%)
calex1	oxide	398		23.60		960	0.278	2.74	1.102	0.888	23.75	0.16	0.6%	23.75	0.6%	23.76	0.7%	24.11	-1.5%
calex1	oxide	398		23.60		1500	0.295	3.05	1.106	0.920	22.92	0.12	-2.9%	23.34	-1.1%	23.35	-1.0%	24.11	-3.1%
calex1	oxide	398		23.60		900	0.291	2.81	1.112	0.894	23.47	0.16	-0.6%	23.57	-0.1%	23.58	-0.1%	24.11	-2.2%
calex1	oxide	398		23.60		300	0.263	2.76	1.099	0.913	23.64	0.28	0.2%	23.67	0.3%	23.67	0.3%	24.11	-1.8%
calex1	oxide	398		23.60		1800	0.291	2.75	1.099	0.892	23.89	0.20	1.2%	23.90	1.3%	23.91	1.3%	24.11	-0.8%
calex1	oxide	398		23.60		1800	0.269	2.74	1.102	0.903	23.75	0.14	0.6%	23.75	0.6%	23.76	0.7%	24.11	-1.5%
Calex1	oxide	398		23.60		1800	0.280	2.76	1.110	0.916	23.35	0.13	-1.1%	23.38	-0.9%	23.39	-0.9%	24.11	-3.0%
Calex	oxide	398		23.60		1200	0.286	2.79	1.111	0.895	23.58	0.15	-0.1%	23.65	0.2%	23.66	0.3%	24.11	-1.9%
STD40	oxide	874		107.40		1800	0.184	2.74	1.084	0.747	107.76	0.88	0.3%	107.76	0.3%	107.76	0.3%	104.44	3.2%
MSSTD-1	oxide	2436	0.64%	156.00	2.89%	14040	0.309	2.74	1.138	0.917	154.15	0.52	-1.2%	154.15	-1.2%	154.36	-1.1%	147.62	4.6%
average													-0.28%		0.00%		0.05%		
CASKL18-98	impure metal	234	0.55%	13.56	2.93%	3600	0.164	2.82	1.047	1.335	14.24	0.06	5.0%	14.31	5.5%	14.30	5.5%	14.18	0.9%
PMA12012	impure metal	329	0.99%	19.37	4.67%	3000	0.325	2.77	1.128	0.673	19.02	0.07	-1.8%	19.05	-1.6%	19.07	-1.5%	19.94	-4.3%
CZA852211	alloy w/Al,3kG DU	1263	0.40%	153.92	1.50%	3600	0.413	3.18	1.183	0.001	152.63	0.38	-0.8%	156.66	1.8%	157.16	2.1%	150.93	4.1%
PMP1482	impure metal	937	0.81%	57.39	3.74%	3660	0.710	3.06	1.322	0.102	56.73	0.14	-1.2%	57.81	0.7%	58.61	2.1%	56.78	3.2%
PMP1487	h.p. impure metal	2417	0.63%	136.62	3.01%	1800	1.116	3.06	1.483	0.030	136.52	0.41	-0.1%	139.12	1.8%	144.03	5.4%	146.47	-1.7%
LM0104	metal	2472	0.75%	404.72	1.84%	13080	1.269	3.10	1.545	0.325	376.63	0.53	-6.9%	384.72	-4.9%	402.62	-0.5%	421.72	-4.5%
PMP12009	h.p. impure metal	2468	0.77%	140.92	3.45%	1800	1.603	3.05	1.664	0.041	134.23	0.51	-4.7%	136.71	-3.0%	146.70	4.1%	149.56	-1.9%
PMA5124	impure metal	2554	0.66%	159.9	3.00%	3600	2.026	3.18	1.785	0.167	126.47	0.40	-20.9%	129.81	-18.8%	143.80	-10.1%	154.77	-7.1%
LM0101	high purity metal	3378	0.78%	368.57	2.63%	3600	2.141	3.32	1.836	0.084	341.97	0.51	-7.2%	353.97	-4.0%	398.22	8.0%	403.67	-1.3%
PMA1485	impure metal	2884	0.74%	171.3	3.43%	3600	2.233	3.04	1.861	0.088	150.88	0.54	-11.9%	153.57	-10.3%	174.17	1.7%	174.77	-0.3%
PMA9391	impure metal	2734	0.37%	160.45	1.92%	3600	2.411	3.00	1.908	0.352	133.94	0.39	-16.5%	136.01	-15.2%	156.74	-2.3%	165.68	-5.4%
LM0093	high purity metal	4190	0.74%	245.20	3.60%	3600	3.225	3.32	2.125	0.275	186.85	0.25	-23.8%	193.40	-21.1%	243.88	-0.5%	253.91	-4.0%
LM0079	impure metal	4074	0.87%	237.00	4.51%	3600	3.863	3.26	2.281	0.335	172.68	0.24	-27.1%	178.09	-24.9%	244.28	3.1%	246.88	-1.1%
average													-9.1%		-7.2%		1.3%		-1.8%
RSD													10.2%		9.9%		4.6%		3.3%

Table II. Multiplicity Results for Impure Items.xls

Sample ID	Sample Type	Reference Pu total (g)	RSD on spec. power	Reference Pu-240eff (g)	RSD on Pu-240 effective	Count Time (s)	Inner/Outer ratio	Sample M	Sample alpha	Pu-240 eff mass (g)	Pu-240 RSD (g)	assay/reference (%)	Pu-240 w/Energy Bias Corr	assay/reference (%)	Strm. Avg. Pu-240eff (g)	assay/reference (%)
PEOF14	h. p. dioxide	1175.3	0.37%	136.83	1.21%	3600	2.96	1.088	0.633	141.40	0.90	3.3%	143.24	4.7%	140.45	2.0%
CASKL1032	MgO crucible	365.0	0.43%	21.92	2.06%	3600	3.09	1.088	0.666	21.75	0.08	-0.8%	22.20	1.3%	22.12	0.4%
PEOF13	h. p. dioxide	1278	0.38%	48.76	2.72%	3600	2.98	1.093	0.782	80.71	0.47	65.5%	81.86	67.9%	77.45	5.7%
CXPROD173	impure dioxide	865	0.48%	52.37	2.26%	3240	2.74	1.064	0.840	53.21	0.36	1.6%	53.21	1.6%	52.42	1.5%
CXPROD173	impure dioxide	865	0.48%	52.37	2.26%	3600	2.76	1.074	0.870	51.78	0.30	-1.1%	51.84	-1.0%	52.42	-1.1%
CASKL10360X	mult. imp. dioxide	996.0	0.58%	61.23	2.75%	3600	2.88	1.151	1.018	59.82	0.28	-2.3%	60.31	-1.5%	60.36	-0.1%
XBLSCL7-98	MSE salt w/Am	199.0	0.63%	11.59	3.16%	3600	3.66	1.057	1.032	11.81	0.05	1.9%	12.48	7.7%	12.06	3.5%
CXLPROD21098C	impure dioxide	737	0.65%	43.31	2.91%	3600	3.08	1.068	1.185	44.15	0.26	1.9%	45.05	4.0%	44.66	0.9%
HRAOXOC21	dioxide w/DU or U	265	0.47%	20.54	2.38%	3600	2.93	1.039	1.540	20.92	0.13	1.9%	21.16	3.0%	16.06	31.7%
FFR12BR	filter residue	607	0.33%	68.42	1.29%	1800	2.91	1.057	1.627	70.46	1.04	3.0%	71.17	4.0%	72.54	-1.9%
FFPFOX021	mult. imp. dioxide	295	0.29%	35.48	1.05%	1800	2.42	1.034	1.674	36.24	0.46	2.1%	35.58	0.3%	35.25	0.9%
XBLC2414	MgO crucible	130.0	0.70%	8.17	4.04%	3600	3.67	1.022	2.453	8.25	0.07	1.0%	8.72	6.8%	7.88	10.7%
XBSOX375A	mult. cont. salt	217.4	0.43%	12.74	2.24%	3600	3.72	1.027	2.926	13.16	0.17	3.3%	13.96	9.6%	13.17	6.0%
RESLC-6	filter residue	80	N/A	9.61	N/A	3600	3.50	1.025	3.624	11.47	0.17	19.4%	12.00	24.9%	9.56	25.6%
XBRCW2188	impure ER salt	493	0.36%	28.77	1.94%	1800	3.42	1.066	3.644	27.21	0.65	-5.4%	28.34	-1.5%	29.88	-5.2%
XBRCW2188	impure ER salt	493	0.35%	28.77	1.94%	14220	3.41	1.071	3.880	25.78	0.23	-10.4%	26.83	-6.7%	29.88	-10.2%
XBS2413	mult.cont.ER salt	521.0	0.30%	30.68	1.85%	3600	4.31	1.074	4.796	26.51	0.57	-13.6%	29.19	-4.9%	31.57	-7.5%
XBS1484	mult.cont.ER salt	337.1	0.44%	19.70	3.10%	2760	4.44	1.047	6.040	17.22	0.48	-12.6%	19.12	-3.0%	20.43	-6.4%
average												-1.6%		1.5%		0.4%
RSD												5.7%		4.5%		10.9%
ASHRCVP83	incinerator ash	276	0.34%	17.50	1.83%	1800	2.66	1.023	6.820	36.31	3.30	107.5%	36.14	106.5%	16.73	116.0%
SCC51A	sand & slag salt	118.7	0.51%	7.39	2.82%	3600	3.13	1.012	7.474	7.66	0.33	3.7%	7.84	6.1%	7.19	9.0%
XBLC12014	MgO crucible	242.2	0.47%	13.77	3.52%	3600	3.45	1.020	7.752	15.20	0.67	10.4%	15.86	15.2%	14.68	8.0%
RESED77F	filter residue	339.0	0.47%	20.23	2.60%	3600	3.60	1.028	10.529	19.41	1.75	-4.1%	20.44	1.0%	20.54	-0.5%
INCA-19	incin. ash w/EU	161	0.35%	16.60	2.02%	14340	3.76	1.015	16.981	17.91	1.84	7.9%	19.04	14.7%	19.24	-1.0%
GVB20-M99	neutron source	2	N/A	0.13	N/A	3600	1.88	1.013	29.269	5.48	1.53	4259.6%	5.22	4051.2%	0.12	4248.3%
GVB17C1	impure dioxide	314		24.61		3600	4.34	1.038	30.108	20.84	9.20	-15.3%	22.99	-6.6%	19.03	20.8%
INCA-19	incin. ash w/EU	161	0.35%	16.60	2.02%	1800	3.75	1.026	30.126	10.25	4.92	-38.3%	10.89	-34.4%	19.24	-43.4%
ARF876595	impure MSE salt	263	N/A	16.53	N/A	5400	3.85	1.021	34.150	11.82	4.60	-28.5%	12.64	-23.5%	15.94	-20.7%
average												-9.2%		-4.0%		-7.5%
RSD												18.8%		18.9%		20.7%

Table III. Multiplicity Deadtime Corrections.xls

Sample ID	Meas. Singles (cps)	Singles Correction Factor	Doubles Correction Factor	Triples Correction Factor	ln(TCF)/ln(DCF)	Meas. Quads (cps)	Quads RSD (cps)	Quads DT-Corr. Factor
calex1	28914	1.003	1.011	1.051	4.547	361	8	1.083
STD40	117820	1.012	1.043	1.206	4.449	649	56	1.349
MSSTD-1	193085	1.018	1.072	1.295	3.718	2461	64	1.512
CASKL18-98	20312	1.002	1.007	1.033	4.654	250	6	1.053
PMA12012	20923	1.003	1.008	1.039	4.801	395	5	1.063
PMP1482	47906	1.008	1.018	1.055	3.001	31196	204	1.089
CZA852211	104662	1.010	1.039	1.151	3.676	4515	62	1.252
PMP1487	120533	1.011	1.045	1.134	2.857	64435	327	1.223
PMP12009	134288	1.012	1.050	1.145	2.775	197060	1306	1.242
PMA5124	151757	1.014	1.056	1.162	2.756	389319	1591	1.272
PMA1485	175479	1.017	1.065	1.166	2.439	2313521	10893	1.279
PMA9391	198369	1.018	1.074	1.204	2.600	787626	2254	1.346
LM0093	288186	1.026	1.109	1.294	2.491	2442244	4352	1.510
LM0079	299050	1.027	1.114	1.314	2.529	4194467	8294	1.548
LM0101	384335	1.035	1.148	1.385	2.360	958009	2890	1.684
LM0104	433283	1.040	1.169	1.474	2.485	205647	815	1.860