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Title: **ASSAY OF SCRAP PLUTONIUM OXIDE BY THERMAL NEUTRON MULTIPLICITY COUNTING FOR IAEA VERIFICATION OF EXCESS MATERIAL FROM NUCLEAR WEAPONS PRODUCTION**

Author(s): **J. E. Stewart, M. S. Krick, J. Xiao, R. J. Le Maire, V. Fotin, L. McRae, D. Scott, and G. Westsik**

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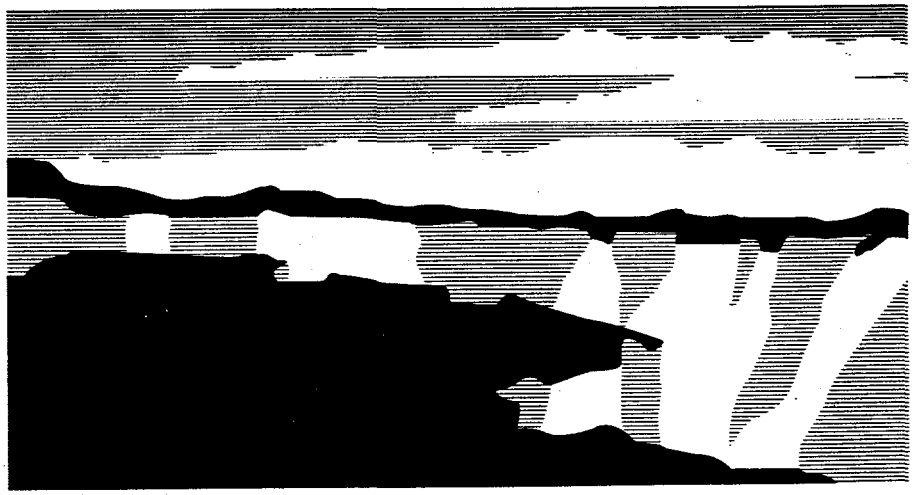

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ASSAY OF SCRAP PLUTONIUM OXIDE BY THERMAL NEUTRON MULTIPLICITY COUNTING FOR IAEA VERIFICATION OF EXCESS MATERIALS FROM NUCLEAR WEAPONS PRODUCTION*

J. E. Stewart and M. S. Krick
Los Alamos National Laboratory
Los Alamos, NM 87545 USA

J. Xiao, R. J. LeMaire, and V. Fotin
International Atomic Energy Agency
Vienna, Austria

L. McRae, D. Scott, and G. Westsik
Westinghouse Hanford Corporation
Richland, WA

ABSTRACT

The US Nonproliferation and Export Control Policy commits the US to placing under International Atomic Energy Agency (IAEA) safeguards excess nuclear materials no longer needed for the US nuclear deterrent. As of January 1, 1996, the IAEA has completed Initial Physical Inventory Verification (IPIV) at the Oak Ridge Y-12 plant, the Hanford Plutonium Finishing Plant, and a plutonium storage vault at Rocky Flats. Two IPIVs were performed at Hanford: one in December 1994 and one in August 1995.

This paper reports the results of thermal neutron multiplicity assay of plutonium residues during the second IPIV at Hanford. Using the Three Ring Multiplicity Counter (3RMC), measurements were performed on 69 individual cans of plutonium residues, each containing approximately 1 kg of material. Of the 69 items, 67 passed the IAEA acceptance criteria and two were selected for destructive analysis.

INTRODUCTION

In December 1994, the International Atomic Energy Agency (IAEA) performed inventory verification measurements on an initial offering of relatively pure plutonium oxide powders and scrap residues at the Hanford Plutonium Finishing Plant. For pure plutonium powder, the IAEA used the standard High-Level Neutron Coincidence Counter (HLNC).[1] For verification of plutonium material containing unknown impurity concentrations, the IAEA used the Three Ring Multiplicity Counter (3RMC) provided by Los Alamos. Measurement results for 21 items representative of the initial offering are reported in Ref. 2. For both powder and scrap, the IAEA used High-Resolution Gamma Spectrometry (HRGS) to verify plutonium isotopic abundances. It was determined during this first Initial Physical Inventory Verification (IPIV) that conventional neutron coincidence counting assay using the HLNC, while producing acceptable results for the pure powder, produced unacceptable results for the scrap items. This is due to unknown levels of (α,n) reactions in the scrap items that arise from impurities.

The second offering, verified in August 1995, included over 600 items, all of which qualified as scrap material. These items were packaged in three nested metal cans, the outer can typically 4 in. in diameter and 7 in. tall. Plutonium masses varied between 800 and 1100 g per item. The measurement methods chosen for inventory verification included the 3RMC, HRGS and destructive analysis (DA). Occasional use was made of facility calorimeters and HRGS systems for comparisons, but not verifications. Sixty-nine items were chosen for verification in the 3RMC. The IAEA had never verified this type of plutonium residue material before.

THE ASSAY SYSTEM

The neutron part of the assay system consisted of the 3RMC, a Canberra 2150 multiplicity Nuclear Instrument Module (NIM), a powered NIM bin, a NIM high-voltage power supply to provide +1680 V for the ^{3}He tubes, an IBM Thinkpad computer, and a Hewlett-Packard 4L

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Laserjet printer. The Canberra 2150 contains the multiplicity counting electronics and provides the +5 V to power the Amptek preamplifiers. The software used for data collection and analysis was the MULTI code, which is a DOS program written specifically for multiplicity counting; the MULTI code is now obsolete and has been replaced by the general NCC code, which is a Windows program written to cover a wide variety of active and passive coincidence-counting applications, including multiplicity counting.

What is now called the 3RMC was originally built in the 1970s as an experimental active well coincidence counter. This counter was converted from an active counter to a passive counter by removing the polyethylene end plugs that held the americium-lithium (AmLi) sources and by replacing them with shorter graphite end plugs. The counter was also upgraded by replacing the 6 original preamplifier boards in the high-voltage junction box with 12 Amptek boards and a derandomizer. The derandomizer accepts the outputs of the 12 preamplifiers and combines them into a single output pulse stream. If any pulses from the 12 Amptek boards overlap, the derandomizer separates them in the output pulse stream, so that there is no dead time in the system following the Amptek boards.

During the conversion and upgrading of this active well coincidence counter, the need arose for a multiplicity counter to be used for an IAEA inspection at Hanford. Because there were no multiplicity counters available and there was not enough time to build or purchase one, the conversion and upgrading of the active well counter was hastily completed; the counter was defined as a multiplicity counter and given the designation 3RMC for the three rings of ^{3}He tubes in the counter.

Because of the history of the 3RMC, its performance specifications are not as good as those of a counter designed specifically to be a multiplicity counter. First, the neutron detection efficiency of the 3RMC is lower than that of other multiplicity counters. The 3RMC has 60 ^{3}He tubes, whereas other multiplicity counters have 80 to 130 ^{3}He tubes. The efficiency of the 3RMC for ^{240}Pu fission neutrons is about 45% compared to a typical efficiency of 55% for other multiplicity counters.

Second, the vertical profile of the 3RMC (i.e., the variation of efficiency with height in the sample cavity) is not as constant as for most other multiplicity counters because the ^{3}He tubes in the 3RMC have a 20-in. active length, whereas most other multiplicity counters have at least a 28-in. active length. Nevertheless, if a six-in.-high assay can is centered in the sample cavity of the 3RMC, the average efficiency over the height of the can is only 0.5% less than the efficiency at the center of the can.

Third, the efficiency of the 3RMC as a function of the neutron energy is not as constant as for most other multiplicity counters. When a counter is designed as a multiplicity counter, the locations of the ^{3}He tubes are carefully selected to keep the efficiency variation with neutron energy as small as practicable. This consideration is important because the multiplicity analysis assumes that all neutrons are detected with the same efficiency. Unless a plutonium item contains a lot of moderating material, the average energy of the spontaneous and fission neutrons leaving the item changes little from one item to the next. However, the energy of neutrons from (α,n) reactions in the plutonium items can vary over several MeV, depending on the item impurities that produce the (α,n) neutrons. Because multiplicity counters are used primarily for the assay of impure plutonium items, their efficiencies should vary as little as practicable as a function of neutron energy.

MEASUREMENTS

The 3RMC was calibrated before the first IAEA inspection in 1994. The parameters needed for multiplicity assay are the electronic dead time, the neutron detection efficiency, the doubles gate fraction, and the triples gate fraction. The first three of these parameters were obtained from the measurement of reference ^{252}Cf sources whose absolute neutron yields are known. The value for the triples gate fraction was obtained from the measurement of plutonium items whose masses and isotopic compositions were known.

Sixty-nine items were selected for assay with the multiplicity counter. For each item the multiplicity analysis determined the effective ^{240}Pu mass from the measured singles, doubles, and triples count rates using the known parameters for dead time, efficiency, doubles gate fraction, and triples gate fraction. Then the plutonium mass was calculated from the effective ^{240}Pu mass using the operator-declared isotopic composition, which was verified by inspector measurements of the isotopic composition.

The measurement procedure was as follows. The items were placed in the 3RMC sample cavity, radially centered on top of an empty can (about 4 in. high) to approximately center the plutonium in the sample cavity. Each item was measured for 20 or 30 minutes. If the assay result was within the acceptance criterion for that item, then the measurement was complete and the result was accepted. If the assay result was outside the acceptance criterion—usually because the measurement had poor

precision as a result of high impurity content—then the item was selected for DA or a longer measurement was made. If a long measurement was made and the assay result was still outside the acceptance criterion, then the item was selected for DA. Sixty-nine items were selected for multiplicity assay and, of those, 67 passed the acceptance criteria and two were selected for destructive assay.

RESULTS

The results presented below are for the 67 items that passed the acceptance criteria. Comments on the two outliers are presented at the end of this section.

It was already known from the first inspection at Hanford that conventional coincidence counting would not be useful for the impure plutonium items in the offering. The data shown in Fig. 1, therefore, are presented only to illustrate the difficulty of applying conventional coincidence counting to this type of material. Fig. 1 shows the doubles rate vs the declared effective ^{240}Pu mass for the 67 assay items; the standard deviation of the doubles rates is negligibly small compared to the scatter. The straight line shown is a least-squares fit to the data. Clearly, assay by the conventional calibration curve technique is out of the question for these items. Neutrons produced by (α,n) reactions with the impurities in the items induce fissions in the plutonium; the resulting fission neutrons then produce coincidence (doubles) counts. Therefore, the doubles rate is dependent on the impurities in the item. For the items in the Hanford offering, the impurities vary widely from item to item.

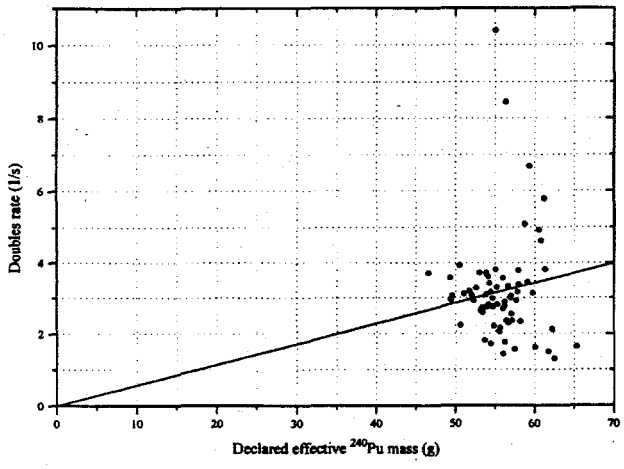

Fig. 1. Doubles rate vs effective ^{240}Pu mass for 67 ARF samples.

Figure 2 shows the assay results for the 67 items using multiplicity analysis; the assay effective ^{240}Pu mass is plotted vs the declared effective ^{240}Pu mass. To avoid clutter, the standard deviations of the assay masses from counting statistics are not shown on this plot. The assay results are presented in Fig. 3 as the percent difference between the declared and assay values vs the declared effective ^{240}Pu masses. The error bars in Fig. 3 indicate one standard deviation from counting statistics; the errors

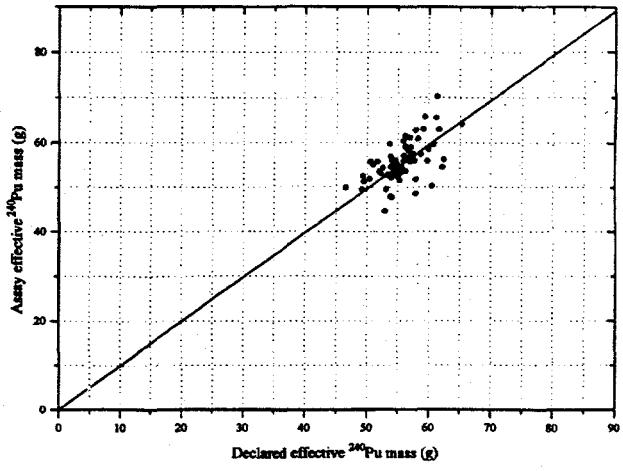

Fig. 2. Assay vs declared effective ^{240}Pu mass for 67 ARF samples.

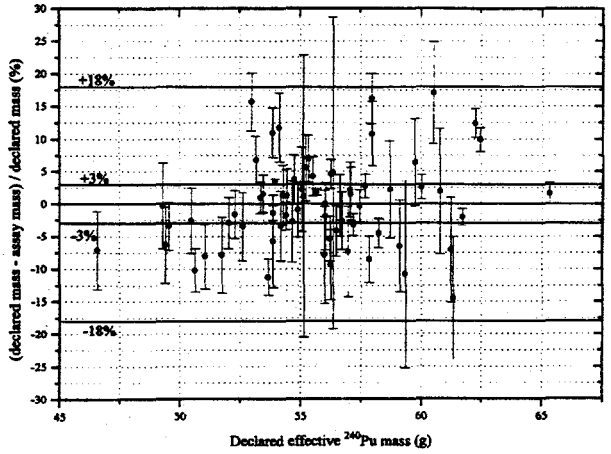

Fig. 3. Difference of declared and assay effective ^{240}Pu masses vs declared effective ^{240}Pu mass.

of the assay masses from counting statistics are dominated by the errors of the triples rates from counting statistics. The scatter of the declared and assay mass differences is comparable to the average standard deviation from counting statistics of the assay masses, indicating that multiplicity assay removes at least most of the assay bias found in conventional coincidence assay of impure plutonium items and that multiplicity assay is limited primarily by the error in the triples rate produced by

counting statistics. There does not appear to be a correlation between the declared and assay mass difference and the declared mass. The ±3% and ±18% limits shown in Fig. 3 are related to the acceptance criteria.

It is important and interesting to look for correlations between the declared/assay mass difference in percent and two quantities: the alpha value and the neutron multiplication. The alpha value is the ratio of neutrons from (α,n) reactions to the neutrons from spontaneous fissions. The neutron multiplication (specifically, the leakage neutron multiplication) is the ratio of the net number of neutrons leaving the item to the number of neutrons produced by spontaneous fissions and (α, n) reactions.

Figure 4 shows the percent difference between the declared and assay masses vs the alpha value; the error bars shown indicate standard deviations from counting statistics only. A typical alpha value is three, meaning that there are three times as many neutrons produced in the item from (α,n) reactions as from spontaneous fissions. There is little, if any, correlation between the percent mass difference and the alpha value, indicating that the variation in detection efficiency with neutron energy is not a major source of bias for the 3RMC and this set of items.

Figure 5 shows the percent difference between the declared and assay masses vs the multiplication; the error bars shown indicate standard deviations from counting statistics only. There is little, if any, correlation between the percent mass difference and the multiplication, indicating the multiplicity analysis model used for these assays is not a major source of bias for this item set. The multiplicity analysis model assumes that the neutron multiplication is the same for all neutrons; this assumption always produces an approximate solution because neutrons originating at the center of an item will—on average— have a higher neutron multiplication than neutrons originating at the surface of the item.

The greatest source of error for these assays is the error of the triples rate from counting statistics. Figure 6 shows the percent standard deviation of the assay mass from counting statistics vs the alpha value for 1200-s measurements, which was the nominal measurement time for the inspection measurements. The percent standard deviation is the measured value and was obtained by the MULTI code as the sample standard deviation of the assay masses from repeat measurements of the same item. There are two reasons for the scatter of the points about the curve in Fig. 6. First, the sample standard deviation

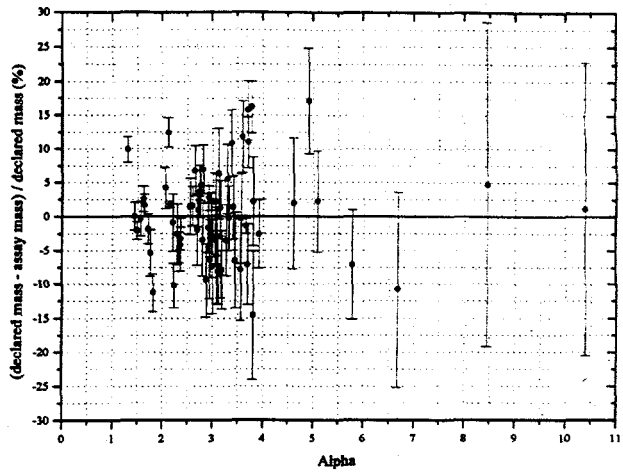

Fig. 4. Difference of declared and assay effective ^{240}Pu mass vs alpha for 67 ARF samples.

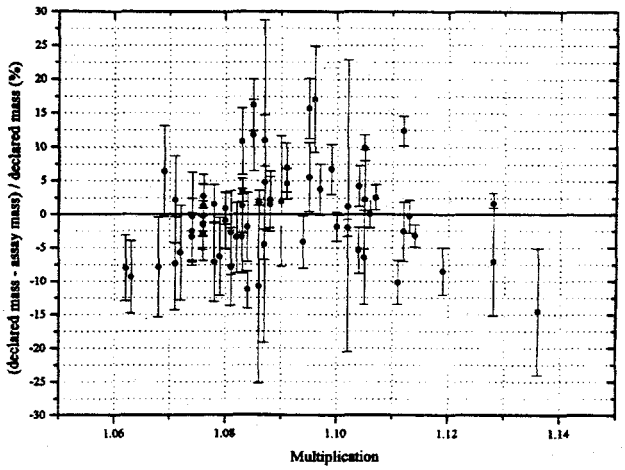

Fig. 5. Difference of declared and assay effective ^{240}Pu effective ^{240}Pu mass vs multiplication for 67 ARF samples.

is itself uncertain because it is estimated typically from six repeat measurements. Second, the standard deviation of the assay mass depends on the plutonium mass and density as well as on the alpha value. The standard deviation increases rapidly as the alpha value increases, so that for a given precision much longer measurements times are required for high-alpha items than for low-alpha items. For example, if the alpha value is 8, the measurement time required for a given standard deviation is about 40 times longer than for an item with the same plutonium mass and density, but with an alpha value of 2.

Of the 67 items that passed the acceptance criteria, 28 assayed within ±3% of the declared masses and all assayed within ±18% of the declared masses.

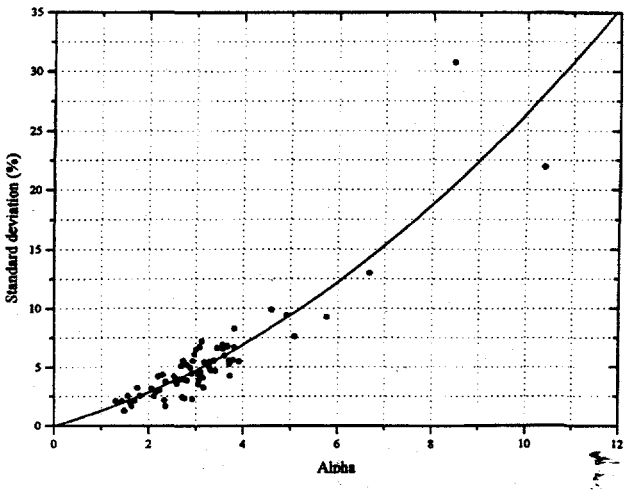

Fig. 6. Percent standard deviation of the assay mass from counting statistics for 1200-s measurements vs alpha for 67 ARF samples.

The average percent difference between the declared and assay masses is 0.02 ± 0.83%.

One of the items that failed the acceptance criteria and that was selected for destructive assay was measured just once for 1200 s. It had a plutonium assay mass difference of 22% relative to the declared mass and assay mass standard deviation of 13%. The alpha value was 5.7 ± 0.8. The failure of this item to meet the acceptance criteria is most likely the result of counting-statistics error, in which case a longer count time would have brought it within the acceptance criterion. The DA gave good agreement with the declared plutonium mass.

The other item that failed the acceptance criteria was remeasured overnight (40 repeat counts of 1100 s each). It had a plutonium assay mass difference of 28% relative to the declared mass and an assay mass standard deviation of 1%, so it was selected for destructive analysis. However, the isotopic composition of the item measured by the inspectors disagreed with the declared values; for example, the declared ^{240}Pu weight percent was 4.94% and the inspector value was 5.81%. The operator also verified that the declared value was low; an operator measurement gave 5.78% for the ^{240}Pu weight percent. The 3RMC measurement data were then reanalyzed with the inspector isotopic values and produced an assay mass that would have met the acceptance criterion for the measurement category for that item—the plutonium assay mass difference was 10% relative to the declared mass with an assay mass standard deviation of 1%. Nevertheless, the assay of this item is biased and needs additional study, because the mass difference is 10 standard deviations and the destructive analysis agreed well with the declared plutonium mass.

COMMENTS AND CONCLUSIONS

The 3RMC played an essential role in the IAEA verification of plutonium residue items offered in both the first and second IPIVs at the Hanford Plutonium Finishing Plant. For the second IPIV in August 1995, the 3RMC was the primary system for mass verification. For the vast majority of items in the second offering, the 3RMC measurements met acceptance criteria, thereby significantly reducing the number of samples taken for destructive analysis. For items with high levels of (α,n) impurities, the 3RMC became precision limited for the standard measurement time of 20–30 minutes. The new Plutonium Scrap Multiplicity Counter (PSMC)[3] has a significantly higher counting efficiency (55% vs 45%) than the 3RMC, and will thus produce better precision in the same counting time. For very highly impure items, e.g., for a values above 10, the method of choice would be calorimetry. These items will require approximately 6 hours of measurement time. The combination of neutron multiplicity counting and calorimetry (both combined with HRGS) will provide quite effective and efficient mass verification for plutonium residue items at Hanford, and will minimize the need for DA sampling and analysis.

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