*Measurements on an Inventory of
Mixed Fissile Materials
in Shipping Containers*

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MEASUREMENTS ON AN INVENTORY OF MIXED FISSILE MATERIALS IN SHIPPING CONTAINERS

by

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ABSTRACT

An inventory contained a large number of previously unmeasured items, many with both uranium and plutonium. We have assembled a suite of instruments and measured the items in a variety of ways. This report first considers the measurements and deduced results in detail before summarizing the important differences with the declarations of the inventory's database.

The appendices referred to in this report are part of a classified version only and are not attached to this unclassified version. The classified report is by the same authors as this report, has the same title (which is unclassified), and is classified as "SRD."

MEASUREMENT GOALS

The general goal of these measurements is to characterize the fissile contents of a set of specific inventory drums. The specific goals are listed below.

- (1) Determine the number of cans bearing nuclear material within each drum.
- (2) Determine the isotopic compositions of the nuclear materials within each drum.
- (3) Determine the total plutonium mass in a drum and the isotopic masses of ^{239}Pu , ^{240}Pu , and ^{241}Pu .
- (4) Determine the total uranium mass and the ^{235}U mass in each drum.
- (5) Perform assays on individual drums in times limited by facility operations (6 hours during the normal working hours; 22 hours during a 24-hour span) and within milestones set to complete the measurements on the inventory.

DIVISION OF LABOR

The instruments' hardware and software were prepared by the Los Alamos National Laboratory (LANL) team to Westinghouse Savannah River Company (WSRC) specifications and installed by both WSRC and LANL personnel.

The instruments were applied to the inventory's drums by WSRC personnel who also used the gamma-ray data to determine the number of cans in a drum and the locations of the fissile

materials for the isotopic measurements. Measurement control data were taken routinely by WSRC personnel. The data analyses for the isotopics, enrichments, and plutonium and uranium masses were done by LANL personnel.

Measurements of the mixed-oxide (MOX) calibration standards were done by LANL NMR-4 personnel (L. Foster, J. Hurd, T. Cremers, and S. Long) prior to shipping the standards to WSRC.

MEASUREMENT PROCEDURES

Segmented Gamma Scanner

The measurements were done with two instruments, each of which operated in two modes. A segmented gamma scanner (SGS) was specially programmed to reach goals (1) and (2). It first performed a vertical scan along a drum and measured the intensity of gamma-rays from a ^{137}Cs source that passed through the drum and any fissile materials. The scan was interpreted by WSRC personnel to give the number of cans in the drum and the heights to use for isotopic analysis. Gamma-ray spectra were then taken at each of these heights for analyses that would give uranium enrichments and plutonium isotopics for each can in the drum. A highly attenuating shutter was in front of the ^{137}Cs source during these measurements, but all the spectra still show a recognizable peak at 661.6 keV from the source; this weakened peak does not interfere with the data analysis and is helpful in resolving questions about certain spectra.

In an ideal world, those drums with small masses of fissile materials would have received the longest count times in the SGS, but it was not practical to arrange this within the working conditions of the facility [goal (5)]. Interpretations of some scans are therefore complicated by poor counting statistics and analyses for isotopics can suffer in precision and accuracy. Fortunately, it is more important to have good accuracy for cans with large masses (hundreds or thousands of grams) of a fissile material, and this is assured by the six-hour (or longer) times used.

Passive-Active Shuffler

Each drum was also placed in a shuffler for neutron measurements to achieve goals (3) and (4). A passive neutron count is related to the plutonium mass in the drum; isotopic information is used in the analysis. This shuffler was not originally designed for passive measurements but it is adequate for the goals of these measurements (although the variation of detection efficiency with neutron energy required knowing the variation for multiplicity counting and a corresponding correction to the count rate). Long count times are needed for drums with small plutonium masses but the several hours used on each drum were generally adequate.

The shuffler in the active mode then measured the count rate of delayed neutrons produced by fissions in the materials after irradiation by neutrons from a ^{252}Cf source. Delayed neutrons are produced by both uranium and plutonium, so the data analysis uses the plutonium mass from the passive count to subtract an estimated count rate caused by the plutonium, leaving an estimate of the count rate from the uranium. The ^{235}U mass is inferred from this net count rate and the enrichment is used to calculate the total uranium mass. The assay time for each drum was 1000 s.

Drums with a small uranium mass and a large plutonium mass might have profited from longer count times, but the small gains in precision would probably have been overwhelmed by the inaccuracies in subtracting the estimated plutonium contribution.

Calibrations

Calibrations of the instruments were done with seven individual cans of MOX (Table I) used in 29 combinations (Table II) within 4 drums thought to be nearly identical to the inventory's drums in capacities and matrices (packing materials). The standards do not span the wide range of materials declared to be in the inventory (in mass, isotopics, physical and chemical forms, or impurities), but they were the best available.

The standards were all very pure and they all had the same total mass (uranium plus plutonium added to approximately 1700 g), the same plutonium isotopics (5.86% ^{240}Pu), and the same uranium enrichment (93.1%); they differed only in the relative amounts of uranium and plutonium.

The calibration drums matched those expected in the inventory. All but the 5-gallon size had one or more cans inside a thick-walled steel pipe surrounded by fiberboard rings. The 5-gallon "drums" (actually "pails") had no matrix; cans were simply placed loosely within them.

TABLE I. CHARACTERISTICS OF THE INDIVIDUAL MOX STANDARDS		
MOX Can ID	U Mass (g)	Pu Mass (g)
SRPSTDPUEU1	1690.552	0.0813
SRPSTDPUEU2	1646.679	43.642
SRPSTDPUEU3	1605.823	87.139
SRPSTDPUEU4	1522.774	174.505
SRPSTDPUEU5	1351.872	348.658
SRPSTDPUEU6	422.443	1308.094
SRPSTDPUEU7	0.1508	1746.842
Pu Isotopics and ^{241}Am Fraction (May 15, 1995)		
^{238}Pu		0.0137%
^{239}Pu		93.853%
^{240}Pu		5.8635%
^{241}Pu		0.2042%
^{242}Pu		0.0659%
^{241}Am		0.0950%
^{235}U Enrichment		93.1109%

TABLE II. COMBINATIONS OF MOX STANDARDS AND DRUM SIZES USED FOR CALIBRATION						
Meas. Number	Drum Size (gal.)	IDs & Positions of Cans			U mass (g)	Pu mass (g)
		Top	Middle	Bottom		
1	55		1		1690.55	0.0813
2	55		7		0.1508	1746.84
27	55		5		1351.87	348.658
28	55		6		422.443	1308.09
13	55	5		6	1774.32	1656.75
14	55	4		7	1522.92	1921.35
15	55	7		6	422.594	3054.94
19	55	6		4	1945.22	1482.6
21	55	5		4	2874.65	523.163
16	55	2	5	6	3420.99	1700.39
17	55	5	2	6	3420.99	1700.39
22	55	5	7	3	2957.85	2182.64
29	55	4	5	7	2874.8	2270.01
3	30	1			1690.55	0.0813
4	30	7			0.1508	1746.84
9	30	5		6	1774.32	1656.75
10	30	4		7	1522.92	1921.35
11	30	6		7	422.594	3054.94
12	30	5		4	2874.65	523.163
18	30	6		4	1945.22	1482.6
20	30	5		4	2874.65	523.163
5	10		1		1690.55	0.0813
6	10		7		0.1508	1746.84
23	10		5		1351.87	348.658
25	10		6		422.443	1308.09
7	5		1		1690.55	0.0813
8	5		7		0.1508	1746.84
24	5		5		1351.87	348.658
26	5		6		422.443	1308.09

DRUM IDS

Three apparent irregularities were found in IDs of drums in the database. They are described here and resolutions suggested.

The database has drum IDs of "B8" and "0B8." Both items have the same Tamper Indicating Device (TID) number, so these items are assumed to be the same drum. The single ID "B8" is used in this report, although the intended ID may have been "0B8."

The database has two pages with the same drum ID of "B9" but with different TID numbers. The declared contents of the two cans are quite different (molten salt button versus enriched uranium oxide). Different TID numbers and different types of materials strongly imply two different drums, so their IDs should also be different. There are two data files generated by

the passive neutron software where the operator typed in drum IDs of “B9” and “0B9,” so these names are adopted here. The passive-neutron results for the plutonium masses match the contents in the database for the two items called “B9.” On this basis, we believe that the drum with the description “molten salt buttons; Pu present, but very high production of neutrons from alpha-n reactions” is the real “B9.” The drum with the description “enriched uranium oxide; assay of zero” is actually “0B9”, not “B9” as shown in the database.

It appears that the drum IDs R8 and S2 have been switched at some time, either when creating the database or when the drums themselves were labeled. Here is the evidence. (a) The numbers of cans in the two drums found from the SGS scans are switched from the numbers in the database. (This is discussed in detail in the next section.) (b) The Pu masses found with passive neutron counting are also switched from those in the database.

NUMBER OF CANS PER DRUM

WSRC personnel interpreted the SGS scans to deduce the numbers of cans and the positions of the fissile materials. The number of cans in a drum deduced from an SGS scan agreed with the number in the database for 83% of the drums. The discrepancies are listed in Table III, with proposed resolutions of the discrepancies.

The spectra and the precision of the deduced isotopics were used to help decide if an apparent can is likely to be a real can or not. These methods and the original scan could be confused by two cans with materials that are exceptionally close together vertically (because of nearly full can or because they are side-by-side in a 5-gal. drum). Additionally, the gap between two cans with intensely emitting contents could appear to be a can itself through the superposition of the tails of the two peaks.

Most of the resolutions in the table favor the database being correct. Here is the list of drum IDs where there may be problems with database: B9, F6, G6, J2, R8, R9, and S2.

TABLE III. RESOLUTION OF DISCREPANCIES IN THE NUMBER OF CANS IN A DRUM

Drum ID	Drum Capacity (gallons)	Number of Cans from the Database	Number of Cans from the SGS Scan	Possible Resolution of the Discrepancy
B7	30	1	3	The spectral lines and the precision of the isotopics indicate the actual presence of three cans. The Pu mass given in the database agrees with the passive neutron assay, so perhaps that single database mass is the sum of all the Pu masses in three cans.
B8	30	2	3	The spectral lines and the precision of the isotopics indicate the actual presence of three cans. Perhaps a page of information on a can was omitted in the database. The Pu mass given in the database agrees with the passive neutron assay, so perhaps the contents of two cans have been merged into one can in the database, leaving two apparent cans in the database.
B9 OB9	30 10	1 1	1 and 2	From the discussion in a previous section on drum IDs, it seems likely that the two database items with the drum ID of "B9" are actually two different drums with IDs of "B9" and "OB9." There are two SGS scans associated with the ID "B9." One scan shows a single can in a 5-gallon pail; the other shows two items in a 30-gallon drum. If these two scans are on two different containers with these nearly identical IDs, there is still some problems. Drum "B9" is said to have a 30-gallon capacity with only plutonium; the gamma-ray spectrum agrees on the presence of plutonium, but the scan shows two cans inside, not one. Drum "OB9" is said to have a 10-gallon capacity, not the 5 gallons read from the scan, containing uranium and a gram or less of plutonium; no spectrum was taken. So the number of cans in "B9" is in disagreement and the size of the container for "OB9" is in dispute.
G2	5	2	1	The declared contents of the two cans are very similar, and the counting statistics on the one can measured are good, so a second can would have been detected in the SGS scan if it were spatially separated from the other can. Cans were placed in 5-gallon pails differently than in the larger-capacity drums, so it is likely that the two cans were at the same height and appeared to be only one can to the SGS scan. We think that the database could indeed be correct in this case.
G3	5	4	2	It is possible that the SGS scan saw only two cans because the four cans were not aligned vertically. If three were nearly on the bottom of the 5-gallon pail and the fourth was resting on top of them, the SGS scan would find two cans. We think that the database could indeed be correct in this case.
H3	5	1	Unclear	The database shows one can containing only U. The scan is difficult to interpret, probably because there are several small items inside, grouped nonuniformly.
H7	5	1	1 or 2	The database shows one can with only 75 g of U. The SGS scan was interpreted as having one can with two objects at different heights. The analyses of the spectra at the two heights show no Pu present at either position. The signal for the SGS scan would have been quite weak from 75 g of U. We think that the database could be correct in this case.
Q9	5	3	1	It is possible that the SGS scan saw only one can because three cans were not aligned vertically. If two were nearly on the bottom of the 5-gallon pail and the third was resting on top of them, the SGS scan would find two cans. We think that the database could indeed be correct in this case.
R3	5	3	1	It is possible that the SGS scan saw only one can because three cans were not aligned vertically. If two were nearly on the bottom of the 5-gallon pail and the third was resting on top of them, the SGS scan would find two cans. We think that the database could indeed be correct in this case.
R8	55	1	3	The three sets of counting statistics are good, indicating that there really are three cans in this drum. From the discussion in a previous section on drum IDs, we believe that the contents of drums S2 and R8 should be switched; drum S2 is said to have three cans, as found with the SGS scan.
R9	55	4	3	The four cans in the database are said to have about the same amounts of Pu, so there is no reason why one of them should be less readily detected than the others. The scan indicates rather clearly that there are only three cans in this drum.
S2	55	3	1	The scan shows only one can in this drum. From the discussion in a previous section on drum IDs, we believe that the contents of drums S2 and R8 should be switched; drum R8 is said to have one can, as found with the SGS scan.
S4	5	3	1	It is possible that the SGS scan saw only one can because the three cans were not aligned vertically. If they were all on the bottom of the 5-gallon pail, the SGS scan would find only one can. We think that the database could indeed be correct in this case.

URANIUM ENRICHMENTS

The ^{235}U enrichments were calculated by the Los Alamos code "PCFRAM" from gamma-ray spectra taken at positions where the SGS scan indicated that cans bearing the fissile materials were located along the vertical axis of the drums. "PCFRAM" uses parameter files that (among many other things) specify the energies at which useful peaks are expected in spectra. One parameter file is the default, but an operator can specify another file to be used for a particular spectrum or change the default file.

Two such parameter files were used here. The first ("LANL_MOX") assumed that both plutonium and uranium peaks will be encountered. This parameter file was generated especially for the MOX standards used in calibration measurements of the instruments. The energy calibration and matrix-attenuation corrections for a spectrum is based strongly on the relative intensities of certain plutonium peaks, so if in fact there is no plutonium the energy calibration may be impossible to form properly and either an error message arises or an improbable interpretation of the uranium peaks is generated. So another parameter file ("Uranium") was used for the cases with no plutonium or only minuscule amounts (e.g., 1 or 2 g).

Regardless of the parameter file applied, there is an important source of uncertainty when dealing with highly enriched uranium (HEU), as exists in much of the materials of this inventory. The amount of ^{238}U in HEU is low, so its gamma-ray peaks are weak and longer count times are needed compared to low-enriched uranium. However, the logistics of doing the measurements on all the drums did not allow for a deliberate matching of count times to the declared materials and it is inevitable that some enrichments will have a loss of precision and accuracy because of this. It also seems that the instrument operators did not always choose measurement positions centered on the fissile material; this resulted in lower count rates than necessary and worse measurement precisions than could have been obtained for the count time.

Furthermore, if the uranium mass is a small fraction of the plutonium mass, the accuracy of the uranium enrichment may be poor because of interferences among the peaks and their tails. Conclusions will be given here about drums with the same type of material (based on the declared material codes), with mention of specific drums when they have noteworthy features.

Material Code 10A, Enriched Uranium Metal with Tritium and Some Possible Plutonium Contamination

Only one of these drums (ID A6) had enough plutonium contamination for PCFRAM with LANL_MOX to even attempt an analysis, but the gamma rays from this small amount were not sufficiently intense for a useful result. With the Uranium parameter file, the deduced enrichments were from 72% to 97%, whereas all the declared are about 93%. The uranium is certainly highly enriched and the declared enrichments are quite plausible.

Material Code 20A, Enriched Uranium and Plutonium

The plutonium provided peaks for use by PCFRAM with LANL_MOX and the measured enrichments are again in agreement with the declared 93% values. The measured enrichments range from 84% to 99% for the six cans in three drums.

Material Code 31B, ZPPR Alloy Metal Pieces

These items have depleted uranium along with plutonium and the results from PCFRAM indicate something less than 0.7%-enriched uranium. For the five items, enrichments from 0.33% to 0.67% were found, compared with the 0.22% declared.

Material Code 40C, Depleted Uranium and Plutonium Scrap

One drum (ID J1) was declared to have only plutonium (despite the material code) and PCFRAM agrees. The other drums contained enough plutonium for analysis and the deduced uranium enrichments are consistent with depleted uranium, although the deduced values for seven cans range from 0.16% to 1.04%.

Material Code 50B, Natural Uranium and Plutonium Alloy

The only drum in this category gave deduced enrichments in its two cans of 0.39% and 1.36%, whereas the declared is 0.71%. The declared enrichment seems likely to be correct.

Material Code 50C, Natural Uranium and Plutonium Compound

The deduced enrichments from the five cans are in essential agreement with the 0.71% of natural uranium; they range from 0.45% to 1.19%.

Material Code 6AP, Enriched Uranium and Plutonium

(Items with material 6AP are split into this and the next two categories because their natures are actually quite different.)

The three cans in this one drum are declared to have 93%-enriched uranium. The deduced enrichments range from 93% to 99%, so they agree.

Material Code 6AP, Molten Salt Buttons (Oxide) and Scrub Alloy Buttons

No uranium is declared for these many drums and none was found with PCFRAM.

Material Code 6AP, Plutonium Oxide (High Fired)

The drum with ID B7 is declared to have 93%-enriched uranium and the measured enrichment is consistent with it.

Material Code 70B, Enriched Uranium and Plutonium Metals, and Alloys

The declared enrichments range from 28% to 93% and the enrichments deduced by PCFRAM track most of them rather well. The worst agreement was with the drum (R6), with the two lowest enrichments, where PCFRAM gave 12%, 10%, and 4% rather than the declared 28%,

28%, and 91%. The masses of uranium in the cans in this drum were all rather low, so the precision and accuracy could be improved. The other drums with higher enrichments and larger uranium masses had less-drastring disagreements.

Material Code 70C, Enriched Uranium Oxide with Minor Amounts of Plutonium (High Fired)

The items in this category have plutonium as a minor constituent; sometimes the plutonium mass was too minor for LANL_MOX and Uranium had to be used. (Other materials with the code 70C are discussed in the two following sections.) The declared enrichments are all 92% or 93% and the deduced enrichments are in quite good agreement.

Material Code 70C, Unirradiated Enriched Uranium (EUN) and Plutonium Nitride (PUN)

Deduced enrichments are again in good agreement with the declared values (near 93%).

Material Code 70C, Enriched Uranium, Plutonium, Zirconium, or Titanium Alloy

These items are declared to have about 93% enrichments. The results with PCFRAM generally agree. An exception is with drum R1 where PCFRAM generates essentially a zero enrichment and the spectrum shows no peaks from any fissile material (although a small peak is seen from the shielded ^{137}Cs source, showing that a spectrum was collected). Both the passive and active neutron measurements (described later) indicate no plutonium or uranium is present. The gamma-ray scan definitely indicated the presence of absorbing material, so there may be titanium or zirconium inside that was thought to also contain uranium when in fact it did not. So the strange enrichment from PCFRAM is another indication that either the declaration for drum R1 is wrong and this drum is actually empty, or the gamma-ray and two neutron measurements were all done improperly. (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)

Material Code 72C, Enriched Uranium Oxide with Plutonium

Many of these cases had plutonium as a minor constituent (1 or 2 g) and this was too little for the parameter file LANL_MOX, so Uranium was used. When the declared enrichment was about 92%, the deduced enrichments generally ranged from 75% to 93%, with clear exceptions of drums P9 and Q1, where the deduced enrichments are substantially lower (20% to 64%) for the eight cans in these two drums. Other declared enrichments were intermediate (20% to 77%). Some deduced enrichments are in excellent agreement with the declarations (e.g., drums H8 and Q7).

Results for some drums (e.g., G7 and H9) must be scrutinized carefully to understand their significance because the drums have multiple cans with some cans having little or no uranium (making the corresponding calculated enrichments appear out of line at first glance when in fact there is too little uranium present to expect a reasonable measurement for the count time available).

Material Code 80B, Plutonium Nitride (PUN) and Plutonium-Thorium Alloy

The amounts of thorium in the three cans of this drum are too small to be important to these nondestructive measurements: 10 g, 1g, <1 g. No uranium is declared for these many drums and none was found with PCFRAM.

Material Code E04, Graphite Scarfings and Fines

No uranium is declared for these many drums and none was found with PCFRAM.

Material Code EAP, Plutonium and Enriched Uranium Oxides

The uranium in these items is a minor constituent, generally a few tens of grams among a few kilograms of plutonium. The deduced uranium enrichments are generally lower than the declared (e.g., 50% rather than 93%), but the uranium peaks are really too weak to give reliable enrichments in the count times used; the ^{238}U peaks are especially weak because there is so little of that isotope.

Material Code ENP, Enriched Uranium, Neptunium, Aluminum Scrap Powder, and Small Pieces from Alloyline

Some of these drums had subgram quantities of plutonium; the agreement between declared and deduced uranium enrichments in these cases when using the parameter file LANL_MOX range from good (drum F9 with 70% and 66%) to poor (drum Q4 with 93% and 67%).

Switching to the parameter file Uranium gave some spectacularly good agreements (drums F7, F8, G6, L6, and Q8), some poor agreements when the uranium masses were low (H7 with 80% and 20%, P8 with 80% and 30%), and agreements of intermediate closeness.

The drums where there seems to be serious disagreements are H7 and P8.

Material Code EOS, Enriched Uranium Oxide Pellets and Powder

Three of these 11 drums have 300 g of declared plutonium and their spectra with LANL_MOX gave uranium enrichments in general agreement with the declaration.

The other drums had no declared plutonium and their gamma-ray spectra supported this. The uranium enrichments with Uranium are in good agreement with most of the declarations in these cases. The most exceptional case is drum J4, with enrichments of 53% and 76% instead of the declared 93%. Drum Q5 is an interesting case in that the enrichments declared for three cans are all different and PCFRAM tracked them fairly well (20%, 30%, and 65% were deduced, compared with the declared 23%, 35%, and 72%).

Material Code MPU, Plutonium Alloy

No uranium is declared for these items but PCFRAM with LANL_MOX gives low enrichments (2% to 25%) with large uncertainties. A visual study of the spectra shows that there

are no genuine uranium peaks; PCFRAM cannot distinguish between statistical fluctuations in the background and genuine peaks.

Material Code PIO, Plutonium Oxide

This solitary drum has no declared uranium but again PCFRAM generates two low enrichments (0.3% and 11%). The uncertainties this time are quite large (about 110%) because no genuine uranium peaks are present, so the deduced enrichments have no meaning.

Results at Various Uranium Fractions

The above results can be compiled in another manner that more clearly shows the performance of PCFRAM on these materials. The parameter of interest here is the ratio of uranium to plutonium.

When there is no plutonium (material codes 10A and EOS), the PCFRAM enrichments are close to the declared enrichments, with some tendency to give results lower than declared values in the 90% range (perhaps because there is so little ^{238}U to create peaks at high enrichments).

When uranium dominates the mass in a can (material codes 70B, 72C, and ENP), the results are also close to the declared values, with some poor agreements for material ENP.

Good agreements are also found when uranium and plutonium masses are about equal (material codes 20A, 6AP, and EOS).

When uranium is a minor constituent, it is also often depleted (material codes 31B, 40C, 50B, and 50C). This weakens the intensity of the gamma rays from ^{235}U and limits the precision and accuracy. Nevertheless, PCFRAM gives enrichments consistent with 0.22%. In other cases (material code EAP) when the declared enrichment is high (93%), PCFRAM tends to give a much lower result (e.g., 50%).

Deduced enrichments when there is no declared uranium (material codes 6AP, 80P, and E04) generally have large uncertainties that indicate their spurious nature. An examination of the spectra shows that no real uranium peaks are present so PCFRAM was performing its mathematical steps on statistical fluctuations in the data.

PLUTONIUM ISOTOPICS

The plutonium isotopics were also calculated by PCFRAM from gamma-ray spectra. The parameter file called LANL_MOX was used in all cases; it was developed from measurements on the seven MOX standards used to calibrate the instruments. The declared isotopics are limited to only the ^{240}Pu fractions and a few cases where combined masses of ^{239}Pu and ^{241}Pu are stated.

If the plutonium mass is a small fraction of the uranium mass, the accuracy of the plutonium isotopics may be poor because of interferences among the peaks and their tails.

Conclusions in this section on the deduced isotopics are again organized according to the type of material (based on the declared material codes). As with the case of uranium enrichment, the precision and accuracy of a result depends on having a long enough count time to clearly form the required peaks in the spectrum. It was not practical to tailor count times to individual drums for optimal results. The time available for a drum was split among the cans in a drum equally, so the count time for a can was small when the number of cans was large.

Material Code 10A, Enriched Uranium Metal with Tritium and Some Possible Plutonium Contamination

No plutonium is declared in these four drums (but contamination is said to be possible), but PCFRAM found information in two cases (IDs A3 and A6) to give crude isotopic estimates. The large uncertainties assigned indicate that they should not be taken seriously, and it is not concluded that plutonium is actually present. A visual inspection of the spectra confirm that very weak but possibly genuine plutonium peaks may be present; these might be caused by plutonium in another drum in the same room or very small contamination amounts within the drum.

Material Code 20A, Enriched Uranium and Plutonium

Each can in this set has hundreds of grams of plutonium, so the spectra are quite clear and the PCFRAM results are in excellent agreement with the declared ^{240}Pu fractions (which are all near 6%).

Material Code 31B, ZPPR Alloy Metal Pieces

The deduced ^{240}Pu fractions are again in excellent agreement with the declared (which range from 9.5% to 11%).

Material Code 40C, Depleted Uranium and Plutonium Scrap

The plutonium masses here are somewhat smaller than in the above cases and generally divided among more cans, so the precisions and accuracies of the PCFRAM results can be expected to be adversely affected. The deduced ^{240}Pu fractions are in general agreement with the declared values, but not with the closeness in the preceding materials.

The worst disagreement is with one of the three cans in drum S2, where PCFRAM gave 33% compared to the declared 6%; but the uncertainty of the 33% is over 100%, so in reality PCFRAM is making no statement about the ^{240}Pu fraction in this case. A visual examination of the spectrum for S2 resolved this problem by showing that no genuine plutonium peaks were even present; PCFRAM had worked with small rises in the fluctuating background. Either this drum is empty or it was not inside the SGS during the count. The background count that is part of the active-neutron measurement was quite high, indicating a large mass of plutonium, so it appears that S2 may not have been in the SGS during the gamma-ray count. (This is still true even if the contents of R8 and S2 are switched; both gave high background count rates, as they should.)

Material Code 50B, Natural Uranium and Plutonium Alloy

The only drum in this category has two cans and their deduced ^{240}Pu fractions closely bracket the declared fractions.

Material Code 50C, Natural Uranium and Plutonium Compound

There is good agreement in this case between the deduced and declared ^{240}Pu fractions, considering that the uncertainties of the deduced values are about 20%.

Material Code 6AP, Enriched Uranium and Plutonium

(Items with material 6AP are split into this and the next two categories because their natures are actually quite different.)

Two of the three deduced ^{240}Pu fractions for these metal pieces are slightly lower than the declared 6%, and the third is slightly higher. But differences are essentially within 2σ and do not seem significant.

Material Code 6AP, Molten Salt Buttons (Oxide) and Scrub Alloy Buttons

The PCFRAM results for these cases have poor precisions. One of the two spectra at different positions of drum B9 has extremely weak plutonium peaks, but this is not true for the other. The ^{240}Pu fractions are consistently smaller than the declared 5.90%, ranging from 2.1% (with an uncertainty >100%) to 4.7% (with an uncertainty of 19%). So statistically, PCFRAM does not disagree with the declared ^{240}Pu fractions and at least agrees that the fractions are low.

Material Code 6AP, Plutonium Oxide, High Fired

Each of these many items is declared to have plutonium in what should be fairly pure oxide forms. The PCFRAM results reflect these favorable conditions. Uncertainties in the deduced ^{240}Pu fractions are generally small (e.g., 4%) and agreements with the declared values (5.5% to 6%) are excellent.

One of the two cans in drum M3 stands away from the others in that the PCFRAM ^{240}Pu fraction is unusually high: $11\% \pm 2.9\%$ instead of the declared 6%. The other can in drum M3 was measured as having the more expected fraction of 5.83%. There is no apparent difference between this single can and the many others, so it seems most likely to be a spurious result.

Material Code 70B, Enriched Uranium and Plutonium Metals and Alloys

Four drums (G5, G9, Q2, and Q3) in this category were declared to have no plutonium and the gamma-ray spectrum support the claim with their very poor counting statistics in the regions where peaks are expected. Plutonium peaks are indeed present, but they are very weak and are unlikely to give very accurate isotopic fractions.

The other five drums have substantial plutonium masses and the PCFRAM ^{240}Pu fractions are in good agreement with the declared fractions.

Material Code 70C, Enriched Uranium Oxide with Minor Amounts of Plutonium, High Fired

The plutonium masses here are very small amounts (1 or 2 g) amidst more than a kilogram of uranium. The signals from plutonium are therefore weak on both absolute and relative scales, giving fairly large uncertainties (40% to >100%) for the PCFRAM results. The best statement on these results is that the deduced ^{240}Pu fractions generally track the declared fractions, considering the large uncertainties.

Material Code 70C, Unirradiated Enriched Uranium (EUN) and Plutonium Nitride (PUN)

Measurement conditions are much more favorable here as the plutonium masses are 100 to 200 g and the uranium masses are reduced. The agreements between deduced and declared values are now quite good.

Material Code 70C, Enriched Uranium, Plutonium, Zirconium, or Titanium Alloy

Plutonium masses in this category are again in the hundreds of grams range, but uranium masses are back to the kilogram range. Some of the plutonium gamma-ray peaks are strong enough to give good statistics and the deduced ^{240}Pu fractions then agree well with the declared. In other cases, the statistics are poor and the agreement is more speculative. But it cannot be said that any firm disagreements are evident.

The strange case of drum R1 was mentioned in connection with the uranium enrichment measurement and will be brought up again in the sections of neutron measurements. We simply repeat here that the gamma-ray measurements agree with the neutron measurements that the drum either has no fissile material (despite the declarations) or the drum was not actually inside the instruments during the measurements. (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)

Material Code 72C, Enriched Uranium Oxide, with Plutonium

Many of these cases had plutonium as a minor constituent (1 or 2 g) or even no plutonium, so the precision in these cases is poor and the large differences between deduced and declared ^{240}Pu fractions cannot be considered significant.

The cases with significant plutonium masses produced very good agreement between pairs of deduced and declared fractions. The ^{240}Pu fractions in these cases varied from 10% to 20% and the PCFRAM results followed them quite closely.

Material Code 80B, Plutonium Nitride (PUN) and Plutonium-Thorium Alloy

This single drum had three cans and, within counting statistics, the deduced ^{240}Pu fractions agree with the declared values.

Material Code E04, Graphite Scarfings and Fines

The PCFRAM ^{240}Pu fraction for the two cans in this drum agrees very closely with the declaration.

Material Code EAP, Plutonium and Enriched Uranium Oxides

A kilogram or more of plutonium dominates the contents of these drums and the good counting statistics in the gamma-ray peaks are the result. The declared ^{240}Pu fractions of about 6% are firmly supported by the PCFRAM fractions.

Material Code ENP, Enriched Uranium, Neptunium, Aluminum Scrap Powder, and Small Pieces from Alloyline

Four of these 15 drums had subgram quantities of declared plutonium, and the others had none. The PCFRAM results agree with this general statement, but in no case are the statistics sufficiently good to give a firm statement about the ^{240}Pu fraction.

Material Code EOS, Enriched Uranium Oxide Pellets and Powder

Plutonium is declared to be absent from all but two of these drums and the results from PCFRAM generally agree. A possible exception is drum H4, where the counting statistics are significantly better than the other cases; some small amount of plutonium may be present. Where plutonium is declared, the deduced ^{240}Pu fractions (6% and 20%) are in good agreement.

Material Code MPU, Plutonium Alloy

These kilogram-plus items (free of uranium) give excellent precisions in the ^{240}Pu fractions and agreements between deduced and declared fractions are excellent.

Material Code PIO, Plutonium Oxide

This solitary drum is declared to have 300 g of plutonium with no uranium. The ^{240}Pu fraction from PCFRAM is in excellent agreement with the declared fraction.

Summary of Plutonium Isotopics Comparisons

There are no clear-cut differences between the declared plutonium isotopics and those calculated with PCFRAM. The differences that do exist can plausibly be attributed to poor counting statistics because of low masses or count times that were too short, both of which lead to small peaks in the spectra and poor precision in the calculated isotopics. When there is a kilogram or more of plutonium, the agreements are excellent.

PLUTONIUM MASSES

Passive neutron data for the drums were analyzed four ways: (1) with a linear calibration curve, (2) with a nonlinear calibration curve, (3) with the known-alpha multiplication correction, and (4) with multiplicity. The results show that the plutonium in the drums has sufficient neutron multiplication and impurity levels to make the first three analyses generally not useful with these

drums. However, for low plutonium masses (less than 15 g declared) the neutron multiplication was assumed to unity and the assay mass was determined from the linear calibration curve; 63 drums met this condition. The rest of the drums were assayed with multiplicity analysis.

In most cases the assay mass exceeds the declared mass, sometimes because the declared mass was probably called zero for lack of a better number. For example, drum Q3 has a declared value of zero but is described as having plutonium contamination; the assay mass is 26 g of Pu. The sum of declared masses of all 63 drums is 65 g; the sum of corresponding assay masses is 271 g. The summed declared mass is probably an underestimate because many small unknown amounts were probably called zero. The summed assay mass is an overestimate because no attempt was made to correct for the effect of neutron multiplication in the uranium. This effect is expected to be small compared to the difference between the declared and assay mass difference. The assay mass of 271 g of Pu is less than 0.2% of the total plutonium mass in all the drums.

The shuffler instrument used for the passive neutron measurements was not designed for multiplicity counting; multiplicity electronics were added to the existing instrument with no other changes. One feature of the shuffler that is particularly bad for multiplicity counting is the single layer of ^3He tubes; detectors designed as multiplicity counters have three to five layers of tubes to obtain a high detection efficiency that is insensitive to the neutron energy. The low detection efficiency was not a major problem for most of these drum measurements because each drum was counted for 6 to 12 hours. However, the strong dependence of the detection efficiency on the neutron energy is an important problem because the energy of the neutrons from the (α, n) reactions in the drums is not known. This energy dependence introduces a bias in the assay results that depends on the neutron energy and the relative number of neutrons from the (α, n) reactions.

In the inventory's drums there is a set of high-fired plutonium oxides with various impurity levels. The difference between the assay and declared masses correlates well with the yield of neutrons from (α, n) reactions, so a correction could be made for the bias in this subset of drums. The same problem also affects the rest of the drums, but a similar bias correction was not possible for them.

For 73 drums (of the 90 containing plutonium), the differences between the assay and declared masses were less than 25%. The total assay mass differs from the total declared mass by $(-0.5 \pm 0.4)\%$. The standard deviation of the 73 differences between assay and declared masses is 10.2%. The 17 drums with differences larger than 25% were studied in more detail, as discussed below.

The 17 drums with assays differing from the declared masses by more than 2σ are now subdivided into five categories, based on the following similarities:

- (1) drum R1;
- (2) drums R8, S2, and S3;
- (3) drums B9, C1, C5, D3, J8, J2, K1, and L4;
- (4) drum G3; and
- (5) drums D4, E8, F3, and S8.

The larger assay-declared mass differences that are statistically meaningful are given in Table IV for this subset of drums. Each category of drums is then discussed.

TABLE IV. DIFFERENCES BETWEEN ASSAY AND DECLARED MASSES FOR DRUMS WITH DIFFERENCES LARGER THAN 2σ .	
Drum ID	(Assay - Declared) / Declared (%)
R1	-100
R8	242
S2	-71
S3	48
J8	39
J2	58
K1	61
G3	-35
L4	221
D4	-27
F3	-27
E8	-49
S8	-55

Drum R1

The declared plutonium mass is 317 g but the assay mass is zero. Either this drum has an incorrect declared mass or it wasn't in the detector when the measurement was made. The active neutron measurement, discussed below, also gave no neutron counts for this drum and the gamma-ray spectrum showed no peaks from fissile material. (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)

Drums R8, S2, and S3

Drum R8 has a declared plutonium mass of 224 g but an assay mass of 765 g. The same masses for drum S2 are 581 g and 168 g, respectively. The IDs of drums R8 and S2 might be reversed (as discussed in the earlier section on drum IDs). If so, the disagreements between the declared and deduced masses are greatly reduced.

The reason why drum S3 has a 48% assay-declared difference is unknown.

Drums J8, J2, K1, C1, D3, C5, B9, and L4

The ratio of neutrons from (α, n) reactions and spontaneous fissions is given the symbol α for convenience. The materials in these drums have α values of about 7 to 50, which are too large for multiplicity assays in this instrument because of large statistical errors and the strong energy dependence of the detection efficiency. These drums simply cannot be assayed by passive neutron counting in this instrument.

Drum L4 is especially interesting because it is supposed to be one of the 4-kg, high-fired oxides, but its count rate is three times larger than the others in the series with material code 6AP (which is well beyond the measurement uncertainty of only about 2%). From the assay data it could be 13 kg of Pu oxide, but it could also be 4 kg of Pu oxide with many times the impurity level of the others in the series. The passive neutron measurement by itself cannot distinguish between these two cases in this instrument.

The active neutron measurement on drum L4, described later, gives a count rate not unlike those from other drums with similar content, except that the background rate is almost five times larger. In fact, this background rate is the same as the singles rate in the passive neutron measurement. A high passive count rate along with a normal active neutron count rate show that drum L4 has an unusual amount of impurities with low atomic numbers undergoing (α ,n) reactions.

Drum G3

There appears to be nothing special about the material in this drum that would account for the -35% difference with the declared mass. The declared mass is rather small, the multiplication deduced from the data is normal, the α value is small (low impurities), and no major matrix materials are listed in the material description. The assay simply does not agree well with declared mass. This may be caused by greater neutron moderation and absorption in this drum than in the calibration drum; the transmission rate (discussed in the section "Uranium Masses") supports this case.

Drums D4, F3, E8, and S8

The assay results with these 55-gallon drums are all less than the declared mass. Drums D4 and F3 have absolute differences only slightly larger than 25%, but the absolute differences for drums E8 and S8 are about 50%. There is no apparent difference among the transmission rates (see "Uranium Masses" section) for these drums and other 55-gallon drums, so the large differences between the assay and the declared masses for drums E8 and S8 are unexplained.

URANIUM MASSES

Determining uranium masses when mixed with plutonium is the most novel aspect of all the measurements on these inventory drums. The shuffler in the active mode irradiates a drum with neutrons from a ^{252}Cf source to induce fission and then counts delayed neutrons emitted after the ^{252}Cf has been rapidly withdrawn into a radiation shield. A measurement on a drum always consisted of 34 of these "shuffles" of the ^{252}Cf source (following a 270-s background count). However, ^{239}Pu fissions as readily as ^{235}U so they both produce delayed neutrons, although a fission of ^{239}Pu produces only about one-third of the delayed neutrons as a fission of ^{235}U . The delayed neutrons from plutonium are an interference because we really want only the count of delayed neutrons from uranium. There is no way to distinguish delayed neutrons from the two elements so a purely computational approach must be used.

The adjustment for the unwanted plutonium contribution to the measured count rate is done by using the plutonium mass deduced from the passive neutron measurement. After subtracting

this estimate rate from the measured rate, an estimate of the count rate from uranium alone is at hand. The calculation of the count rate from plutonium involves calibration parameters estimated from measurements on standards, in this case the seven MOX standards in assorted configurations.

Based on the experience with the MOX standards, the measured delayed neutron count rate r is taken to be this function of the ^{235}U mass m_U and the ^{239}Pu mass m_{Pu} .

$$r = k_1 m_U^2 + k_2 m_{Pu}^2 + k_3 m_U + k_4 m_{Pu} , \quad (1)$$

where the four k 's are the calibration parameters. There is a set of parameters for each drum size. The second-order polynomial form of Eq. (1) allows for self-shielding effects through negative values for k_1 and k_2 . There is no direct expression of any interaction between the uranium and plutonium. Monte Carlo simulations and experience with the MOX standards agree that these interactions are minor because multiplications are generally close to unity. Interactions among materials in the cans within the same drum have similarly been shown to be negligible because even when cans are stacked on top of each other the fissile materials are generally separated by large air gaps. The simple formula in Eq. (1) describes the measured count rates from the MOX standards quite well.

Equation (1) yields the ^{235}U mass through this quadratic equation.

$$m_U = \{-k_3 \pm \sqrt{k_3^2 - 4 k_1 (k_2 m_{Pu}^2 + k_4 m_{Pu} - r)}\} / (2 k_1) . \quad (2)$$

The values of k_1 and k_2 are generally negative (representing self-shielding) so there will be a set of low masses and another set of high masses that give the same count rate. But in practice the mass resulting from use of the minus sign before the radical is unreasonably large (tens of kilograms or more) whereas the plus sign gives masses close to the declared masses (zero to a few kilograms, according to the drum). The positive root is also the one that gives the correct masses for the calibration standards. The total uranium mass in a drum is the ^{235}U mass divided by the enrichment fraction.

When there is no uranium mass present in a drum, Eq. (2) would ideally produce exactly zero, but this is unlikely to happen because of statistical uncertainties in the count rate r , small errors in the plutonium mass m_{Pu} , and calibration errors in the four k parameters. The difference between r and the m_{Pu} terms should be small (instead of exactly zero), but k_1 is likely to be even smaller so the two resulting values of m_U are likely to be very negative and very positive; in other words, two impossible masses are a signal that the uranium mass is consistent with zero.

A small mass of ^{235}U combined with a large mass of ^{239}Pu is difficult to retrieve accurately with Eq. (2) because the term $(k_2 m_{Pu}^2 + k_4 m_{Pu} - r)$ is a small difference between two large numbers; the statistical uncertainty in r and the calibration uncertainties in k_2 and k_4 limit the accuracy of this small difference. When the plutonium mass is nearly zero, this same term is almost $-r$, making Eq. (2) much better at generating an accurate ^{235}U mass.

Uncertainties in the masses are calculated by standard error propagation techniques, including random errors from counting statistics and systematic errors in the four k parameters. Equation (1) is a fairly accurate expression for the count rates from the MOX standards, so the uncertainties in the k 's are fairly small and probably are underestimates for use with the wide variety found in the inventory itself.

Another potential source of bias are differences between the matrices in the inventory drums and the drums used for calibration with the MOX standards. The shuffler has flux monitors that count neutrons during the irradiation of a drum by the ^{252}Cf source. The flux monitors are sensitive to the amount of neutron-energy moderating material (primarily hydrogen) in the matrices and may be used to adjust the measured count rates for differences in moderation within different drums. The flux monitor responses were used in these measurements to look for differences among the calibration standards drums and the inventory drums. There is no strong difference between the inventory drums and the standards drums (implying that the packaging of all the drums was very similar), so the adjustments formed from these data did not improve the assay results. Therefore, flux monitor data were not used in calculating the uranium masses reported here.

There are a few transmission rates that raise questions about the corresponding drums. Are drums with IDs of A3 and A4 really of the 55-gallon size? Their transmissions are more in keeping with the 30-gallon size, or else their packaging is different from the other 55-gallon drums. Similar questions can be asked about drums E9, B9, and H9. In these cases they are all declared to be 30-gallon drums but appear to be more like the 5- and 10-gallon sizes, or else they are 30-gallon drums with unusual packagings.

More important differences among the various drums' contents are caused by the differences between the inventory fissile materials and the calibration MOX. The MOX standards include only the high-mass range (1700 g or more), they all have the same uranium enrichments (93%) and the same plutonium isotopics (5.86% ^{240}Pu), and there are essentially no impurities that could undergo (α, n) reactions or affect neutron transport. The inventory materials have masses from subgram to several kilograms, enrichments and isotopics range widely, and important impurities are often detected in the passive neutron counts. There is little hope of accurately adjusting active-neutron count rates for all of these effects when the true natures of the fissile materials are incompletely known. Even when it is likely that a drum contains fissile material in a rather pure metal form, the shape of the metal is not well known and cannot be inferred from the shape of the can (unlike a powder).

An attempt at a partial adjustment was made by simulating the active-neutron process with a Monte Carlo code (MCNP) for both the MOX standards and models of the types of materials in the inventory. The intent was to adjust the measured count rates so that they would then fall on the calibration "surface" of Eq. (1). It was found that in fact the adjustments were more likely to lead to a uranium mass farther from the declared mass than the unadjusted count rate. So the Monte Carlo approach was abandoned as too ambitious for this inventory with its diversity and large differences with the MOX standards. The count rates were not adjusted by either a function of the flux monitor count rates or by Monte Carlo simulations. The count rates used are simply the measured rates with background subtractions and small adjustments for the slow decay of the ^{252}Cf source over the course of the measurements.

While calculating the uranium masses from Eq. (2), the plutonium masses used are those found from the passive-neutron multiplicity assays. Any difference of a plutonium mass from the true value has an effect on the uranium mass for that drum, but with only about one-third the impact; an error of 0.9% in m_{Pu} , for example, produces an error of only 0.3% in m_{U} .

Generalized results grouped by material code are discussed in the following sections. The first several material types are metals (often with very small amounts of uranium) and the assay results are poor, for reasons given in the appropriate sections.

Material Code 10A, Enriched Uranium Metal with Tritium and Some Possible Plutonium Contamination

Ingots of fissile materials produce lower count rates than oxides because of their greater self-shielding. Count rates will increase as the thickness of the metal decreases, and then may eventually decrease again if the thickness continues to decrease. Since nothing is known about the geometric forms of these metal pieces, it is difficult to draw conclusions about differences in deduced and declared uranium masses. The sum of the assay results for these four drums is 4.4% lower than the sum of the declared masses. For the individual drums there are differences with the declared masses of -5%, +17%, -17%, and -43%. There are obviously amounts of ^{235}U present on the order of a kilogram in each drum, as declared.

Material Code 20A, Enriched Uranium and Plutonium

These are declared to also be unspecified metal pieces, but now with plutonium as well as uranium. The sum of the assay results for these drums is 34% higher than the sum of the declared masses. The individual drum differences between the deduced and declared uranium masses are now 4%, 44%, and 91%, but it cannot even be said if the 91% difference is a real difference without knowing more about the nature of the metal pieces.

Material Code 31B, ZPPR Alloy Metal Pieces

The geometric form of these pieces is somewhat moot in that the uranium masses are much less than the plutonium masses. The uranium is depleted, placing it further from the calibration range. The deduced uranium masses are several times larger than the declared, but this cannot be interpreted as identifying problems with the declared masses because the total ^{235}U mass is only 24 g.

Material Code 40C, Depleted Uranium and Plutonium Scrap

Although three of these four drums are said to contain powders instead of metals, one has no uranium and the others have depleted uranium. The result with the drum devoid of uranium (J1) is consistent with the declaration, but the deduced masses for the other three have the same problem as in the previous case: there is too little uranium (10 g of ^{235}U) mixed with large amounts of plutonium for an accurate uranium assay.

Material Code 50B, Natural Uranium and Plutonium Alloy

The one drum in this category apparently has a metal alloy of natural uranium and plutonium, with the plutonium mass again being the larger of the two. The declared ^{235}U mass is only 16 g. The presence of uranium is confirmed, but the assay value cannot be taken as accurate.

Material Code 50C, Natural Uranium and Plutonium Alloy

These two drums are much like the previous case (including the natural uranium), but now one drum is declared to have only 0.1 g of ^{235}U while the other has 684 g (and dominates the 471 g of plutonium declared for this drum). The first case is quite hopeless but in the second case (S1) the assay value is “only” 35% lower than the declared. These results at least concur that the one drum has no, or almost no, uranium, while the other drum has uranium close to the declaration. The sum of the assay values is 65% higher than the total declared mass.

Material Code 6AP, Enriched Uranium and Plutonium, Molten Salt Buttons (Oxide), Scrub Alloy Buttons, and Plutonium Oxide (High Fired)

All of these many drums’ diverse contents are without uranium except for one (B7), which is declared to have both uranium and plutonium. For this uranium-bearing drum, the assay mass is 85% higher than the declared, mostly likely because the materials are metals and widely different in geometric form from the MOX powder standards. So the contents of drum B7 are consistent with the measurements.

Four other drums in this category have molten salt or scrub alloy buttons with no declared uranium and hundreds of grams of plutonium. All of the assay results show hundreds of grams of ^{235}U , most likely because these metals differ so widely from the MOX standards.

The remaining 26 drums are said to have plutonium oxide without uranium and indeed the results from Eq. (2) concur. The negative root gives large negative masses while the positive root gives large positive masses. The count rate is entirely from the plutonium, so subtracting a count rate estimated from the plutonium mass is difficult to do accurately because the two count rates are both large; a small percent error in one results in large positive and negative uranium masses.

Material Code 70B, Enriched Uranium and Plutonium Metals and Alloys

More metal pieces are in these drums but now the uranium masses generally dominate. Two deduced masses agree very well with the declarations (drum F5 with an 8% difference and drum R6 with essentially no difference), but it is difficult to give them much significance in light of the previous experiences with metals. Results with the other seven drums show the usual large differences with the declared masses, but at least indicate the presence of large amounts of uranium.

Material Code 70C, Enriched Uranium Oxide with Minor Amounts of Plutonium (High Fired), Unirradiated Enriched Uranium and Plutonium Nitride, Alloy, and Enriched Uranium Oxide (with Plutonium)

These materials are enriched uranium oxides, often (but not always) with small and negligible amounts of plutonium. These materials are somewhat similar to the MOX standard of 1700 g of enriched uranium and practically no plutonium, but the uranium masses have a wide range. This is an especially important material type because it contains 28% of the total ^{235}U of the inventory.

For the many drums with less than a few grams of plutonium, the deduced uranium masses are consistently 30% to 50% higher than the declared. This could be easily a bias introduced by

the limited calibration. But no one of these drums stands out as dramatically different from the others, so the declarations are self-consistent.

Three drums have roughly equal mass of the two elements and the deduced masses are roughly equal to the declared (ranging from 12% low to 64% high).

When the plutonium mass is not insignificant, the deduced uranium masses are more widely scattered relative to the declared masses. At best, they indicate the presence of large amounts of uranium, but quantitatively they seem suspect.

Drum R1 was mentioned in the passive neutron section as either being empty or not present in the assay chamber during the passive assay. The same can be said for it from the active neutron interrogation. It produced no delayed neutrons despite its declaration of hundreds of grams of ^{235}U . (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)

The sum of the assay results is 16% higher than the sum of the declared masses, so for this important material the agreement is fairly good.

Material Code 72C, Enriched Uranium Oxide with Plutonium

Some of these MOX powders have insignificant amounts of plutonium, but the uranium assays are about 50% larger than the declared amounts. One drum has contents more similar to the calibration standards than most (950 g ^{235}U , 95% enriched, 350 g Pu); in this case the measured ^{235}U mass is only 3% higher than the declared. This material type contains 16.5% of the inventory's total uranium.

The sum of the assay results is 37% higher than the sum of the declared masses.

Material Code 80B, Plutonium Nitride and Plutonium-Thorium Alloy

No uranium is declared for this one drum and the data analysis is consistent with this statement.

Material Code E04, Graphite Scarfings and Fines

No uranium is declared for this one drum and the data analysis is consistent with this statement.

Material Code EAP, Plutonium and Enriched Uranium Oxides

These MOX materials have uranium as very minor fractions of the fissile material. The small count rate from the uranium is impossible to separate cleanly from the stronger rate from plutonium. The small count rate is the difference between two large numbers, so any relatively small errors in the large numbers results in a relatively large error in their small difference. In many of these cases the differences were negative, which is not surprising under these conditions, and shows that any uranium present has much less mass than the plutonium.

There are two cases (drums K7 and M7) where the uranium mass is large enough to be detected. The differences with the declared uranium masses are 56% and 15%.

Material Code ENP, Enriched Uranium, Neptunium, Aluminum Scrap Powder, and Small Pieces from Alloyline

Now the declared plutonium mass is negligible, but the physical form is a mixture of powder and pellets with no indication of how much oxide is in the two forms. The assay results are generally twice as large, or larger, than the declared masses, probably because most of the uranium is in the form of pellets which are more dense than the oxide powders used for the calibration. But the results clearly indicate the presence of the uranium in fairly large masses.

The total declared mass is 15% higher than the sum of the declared masses. Considering the difference in physical form of these materials from the calibration standards, this difference is not large.

Material Code EOS, Enriched Uranium Oxide Pellets and Powder

Some of these mixtures of pellets and powders of enriched uranium have plutonium and others do not; when plutonium is present, it is an important fraction of the total fissile amount. Once again there is a nearly uniform trend for the deduced uranium mass to be larger than the declared mass by 5% to 100%, with better agreement occurring when plutonium is also present. This material type has 12% of the inventory's declared ^{235}U .

The sum of the assay masses is only 6% higher than the sum of the declared masses, so perhaps the pellets are a minor portion of the material in these drums and the calibration standards are especially valid here.

Material Code MPU, Plutonium Alloy

No uranium is declared for this one drum and the data analysis is consistent with this statement.

Material Code PIO, Plutonium Oxide

No uranium is declared for this one drum and the data analysis is consistent with this statement.

Overview of Uranium Mass Results

The only basis for evaluating the accuracy of the deduced uranium masses is a comparison with the declared masses, but the accuracy of the declared masses is unknown. An agreement of the deduced and declared masses implies that both are probably accurate. A disagreement in the two masses does not indicate which one is in error or if either is accurate. But this comparison is the best that can be done at this time.

Table V summarizes the comparison for the 102 drums that had uranium. Shown in this Table are the percent of the drums with mass differences within a percentage bin (column 2), the cumulative percent of the drums (column 3) with mass differences less than the upper limit of each bin, and the cumulative percent of the uranium masses (column 4) less than the upper limit of each bin. The fraction of the drums in a bin is fairly constant (at about 8%) until the 70% difference bin

is reached (column 2) where the fraction drops until the last bin. About 75% of the uranium mass in this inventory is below the 70% difference bin, implying that drums with larger masses have smaller percent differences. This is seen more easily from the other columns in Table V. About 18% of the cases have less than 20% differences (column 3); however, about 30% of the mass has less than 20% difference with the declared (column 4).

It is a general rule that the drums with larger uranium masses have the smaller differences with the declared values. All of the drums with the smallest uranium masses (under 350 g) have the largest differences; drums with the material codes 31B, 40C, 50B, 50C, 70C, and EAP have especially small uranium masses so even small absolute errors in the masses will lead to large percent errors. Although almost 22% of the drums have differences greater than 100%, only 7% of the total uranium mass is in these drums. This shows that the main reason for the large differences here is that the uranium masses in these drums are small; both the declared and deduced masses are likely to have large percent errors.

The most common result for a deduced uranium mass is an overestimate compared to the declared mass. Assuming the declared masses are accurate, this is likely to be caused by the incompatibilities of the materials with the limited MOX standards and also by having small uranium masses mixed with large masses of plutonium.

An obvious mismatch with the calibration standards occurred with the drums assigned material code ENP. Although this material is oxide with plutonium as a minor constituent, the physical form is pellets (with some powder) with a much larger density than the powder in the calibration standards. The resulting assays for the uranium mass are generally quite different from the declared values and in some cases no masses could even be calculated.

TABLE V. COMPARISON OF DECLARED AND DEDUCED URANIUM MASSES			
Upper Limit Percent Difference	Percent of the Drums Between this Upper Limit and the Next Higher Limit	Cumulative Percent of the Drums Within the Upper Limit	Cumulative Percent of the ²³⁵ U Mass Within the Upper Limit
10%	9.8%	9.8%	19.6%
20%	7.8%	17.6%	29.8%
30%	6.9%	24.5%	37.9%
40%	10.8%	35.3%	54.3%
50%	8.8%	44.1%	65.5%
60%	9.8%	53.9%	76.3%
70%	3.9%	57.8%	80.2%
80%	2.9%	60.8%	82.8%
90%	4.9%	65.7%	90.1%
100%	3.9%	69.7%	93.1%
>100%	21.6%	100.0%	100.0%

The presence of large masses (hundreds to thousands of grams) of uranium is clearly indicated by the active neutron method after a subtraction for the plutonium contribution to count rates, but the accuracy of the deduced uranium masses is difficult to assess.

To test the appropriateness of Eq. (1), a more general form of Eq. (1) was fitted to the inventory's data, as follows:

$$r = k_1 m_U^{n_1} + k_2 m_{Pu}^{n_2} + k_3 m_U^{n_3} + k_4 m_{Pu}^{n_4} . \quad (3)$$

The eight parameters in Eq. (3) could not be evaluated from the limited number of calibration standards available in this study, but the inventory has more than enough data to justify eight parameters. The best-fit values of the k and n parameters were close to those used in Eq. (1), but the subtle differences had some interesting effects. The only large changes in the k parameters were k_1 for 30-gallon drums, k_2 for 55-gallon drums, and k_2 for 30-gallon drums. The values of n_1 and n_2 ranged from 1.89 to 2.18, but stayed close to the 2.00 assumed in Eq. (1). The values of n_3 and n_4 ranged from 0.92 to 1.08 compared to the 1.00 assumed in Eq. (1). Values of the parameters n similar to these would have presumably been found with more extensive calibration, but clearly the simple values in Eq. (1) are justified by this exercise. Uranium masses from Eq. (3) are especially better than those from Eq. (1) when high-fired oxides are involved (the inventory material most similar to the calibration standards).

The more general relation in Eq. (3) has the mathematical disadvantage that there is no closed-form solution like Eq. (2), but it is not difficult to solve it numerically. While negative values for m_U or square roots of negative numbers can be generated by Eq. (2), this cannot occur with numerical solutions to Eq. (3). In future work of this sort, Eq. (3) should be given consideration instead of Eq. (2).

SUMMARY OF APPARENT ANOMALIES

Some differences with the declarations in the database were found in all categories of this study. They occur in only a small fraction of the inventory and many are quite minor. But a few are significant and may be important enough to warrant further measurements specially devised for them. These apparently significant cases are listed now to point them out clearly; they are also described below among the other cases.

- R1; this drum appears to contain no uranium or plutonium, but there is some dense material that absorbs gamma rays. (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)
- R8 and S2; the IDs of these drums may be switched. If not, there are large disagreements between the declared and measured fissile masses.
- 0B9 and B9; the database has two drums called B9 and our assumed names of 0B9 and B9 may be incorrect.
- R9; there may be three cans in this drum instead of four.
- G3; there is an important disagreement between the declared and measured plutonium masses.
- S3; there is an important disagreement between the declared and measured plutonium masses.

- E8; there is an important disagreement between the declared and measured plutonium masses.
- S8; there is an important disagreement between the declared and measured plutonium masses.

It should be noted that it was impossible to measure the plutonium contents of eight of the drums because the neutron background rate was too high, apparently from (α ,n) interactions with unusually high levels of impurities. The drum IDs with this problem are listed in the “Plutonium Masses” section below.

The complete list of anomalies is now given in the following sections. Those given in the above list are repeated in the complete list.

Drum IDs

There is understandable confusion between the two items below, arising from IDs that have leading zeros. Leading zeros in ordinary numbers are of no importance and are normally not shown, but they seem to be very important to the IDs here and cannot be dropped.

- The inventory database shows drums with IDs of 0B8 and B8, but they have the same TID. We assume these are actually two cans in the same drum. Only the drum ID B8 is used in this report.
- The inventory database shows two drums with IDs of B9 that have different TIDs. We assume that these are indeed different drums and use the IDs of 0B9 and B9.

The next observation is more speculative, but the measurement data are much more consistent with the declared contents if indeed two IDs have been inadvertently switched at some time in the past.

- From the results of the neutron measurements, we suspect that the IDs R8 and S2 have been switched; otherwise, the deduced contents of these two drums do not match declared contents very well.

Drum Sizes

The transmission and reflection of neutrons from the ^{252}Cf source are measured with flux monitors. The count rates are generally neatly grouped according to the drum sizes, apparently because the packaging of the drums of each size is very similar; but there are a few anomalies. The true sizes of drums could be readily determined by simple observation, but the relative inaccessibility of the inventory has not made this possible at this time.

- Drums A3 and A4 are said to have 55-gallon capacities. From the neutron transmission measurements they appear to be 30-gallon drums. If in fact they are 30-gallon drums, their packaging must be different from the others of that size.
- Drum E9 is said to have a capacity of 30 gallons. From the neutron transmission measurements it appears to be a 5-gallon drum. If in fact it is a 5-gal. drum, its packaging must be different from the others of that size.

- Drums B9 and H9 are said to have 30-gallon capacities. From the neutron transmission measurements they appear to be 10-gallon drums. If in fact they are 30-gallon drums, their packaging must be different from the others of that size.

Number of Cans Per Drum

The number of cans is deduced from peaks in the vertical gamma-ray scan done with the SGS. In some cases, there is room for different interpretations of what constitutes a genuine peak. This is one explanation for differences in the deduced number of cans with the database. Another explanation is that the database has errors.

The number of cans in drums R8 and S2 differ from those in the database. But if (as we believe) the IDs are switched, the numbers of cans in these drums agree with the database information.

Drum R9 is declared to have four cans with nearly equal amounts of plutonium, but only three cans were apparent from the gamma-ray transmission scan.

Pu Isotopics and U Enrichments

From the gamma-ray measurements, a few anomalies were found.

- The spectrum from drum S2 shows no indication of uranium or plutonium. Either this drum is empty or it was not in the SGS during the count. A large background count rate during the active-neutron measurements shows that plutonium is in this drum, so apparently the gamma-ray spectrum was taken without the drum in the SGS or there is strongly-attenuating material inside.
- There is one drum for which we have no gamma-ray spectral data; its ID is J1. So while this is not strictly an anomaly, it is noted here as an incomplete case.
- PCFRAM uranium enrichments for drums R6, H7, P8, and J4 are much lower than the declared and either a true discrepancy is indicated or the measurement-analysis process failed in this case.
- The spectrum from drum R1 has only a peak from the ^{137}Cs transmission source. No plutonium or uranium is indicated. Either the drum has no fissile material, the fissile material is completely attenuated, or the drum was not in the SGS during the measurement. (After this result was obtained, the drum was opened and in fact the contents did not include any uranium or plutonium.)

Plutonium Masses

These are observations from the multiplicity analysis of the passive neutron data.

- The following drums had amounts of impurities so large that passive-neutron analyses could not be done because of the high neutron production rates from (α, n) reactions: B9, C1, C5, D3, J2, J8, K1, and L4.

These are not anomalies, but they are listed here to record the fact that they could not be measured.

- Drum L4 either has 13 kg of relatively pure oxide or 4 kg with more than the normal amount of impurities. (Uncertainties on these values are about 2%, which are much less than the difference between them.) The active-neutron data indicate a high level of impurities, so the second option is more likely.
- The plutonium assay for drum G3 is 35% less than the declared value, an unusual situation among all the drums measured. There is no apparent reason for a false value, so the declaration may be in error.
- The plutonium assay for drum S3 is 49% higher than the declared value. There is no apparent reason for the assay to be false, so the declaration may be in error.
- The plutonium assays for drums E8 and S8 are significantly lower than the declared masses. There are no apparent reasons for these differences, so the declared values may be in error.
- The data for drum R1 indicate that no plutonium is present, despite the declaration of 317 g. The active-neutron measurement also shows no uranium, whereas 75 g of ^{235}U is declared, and the gamma-ray spectrum showed no indication of any gamma-ray-emitting material. Either the declarations are wrong or the drum was not in the shuffler during these measurements. (After this result was obtained the drum was opened and in fact the contents did not include any uranium or plutonium.)
- No plutonium is declared for 30 of the drums, although 18 of them mention plutonium contamination. All but a few of the drums have statistically significant coincidence rates consistent with the presence of plutonium. Several of the 12 drums that have neither a declaration nor a mention of any plutonium have significant coincidence rates equivalent to grams of plutonium. Table VI lists these drums in decreasing order of their measured plutonium masses. It is difficult to say (without remeasurements) if all of these apparent masses are real or if some were caused by fissions in uranium, spontaneous fissions from other elements, or cosmic-ray events (which is unlikely because the normal background rate from the shuffler's ^{252}Cf sources and cosmic-ray events was a very low 5 counts/s).

TABLE VI. APPARENT PLUTONIUM IN DRUMS WITH NO DECLARED PLUTONIUM MASSES			
	Declared Pu Contamination	Assay Pu Mass (g)	σ of Pu Mass (g) ^a
Q3	²³⁹ Pu	25.770	0.092
P9	²³⁹ Pu	25.08	0.13
Q1	²⁴² Pu	18.041 ^b	0.034
H4	none	14.592	0.031
G5	Pu	14.032	0.053
L5	none	11.443	0.041
F8	Pu	7.205	0.028
E9	none	5.637	0.031
K2	none	4.850	0.018
H3	Pu	4.08	0.15
R5	Pu	2.853	0.040
Q2	²³⁹ Pu	2.188	0.06
G6	Pu	1.629	0.022
G9	Pu	1.337	0.010
L6	Pu	0.58	0.021
H7	Pu	0.573	0.020
40	none	0.463	0.008
F7	Pu	0.459	0.015
P8	Pu	0.392	0.016
H6	none	0.383	0.006
J6	none	0.358	0.006
59	none	0.313	0.008
C4	Pu	0.296	0.011
J4	none	0.224	0.010
Q8	Pu	0.207	0.006
B4	Pu	0.175	0.007
A3	none	0.147	0.005
A4	none	0.128	0.009
A6	none	0.010	0.004
A8	none	0.000	0.006

a. The σ are uncertainties from counting statistics.

b. The database says that ²⁴²Pu may be present. To calculate this Pu mass, it was assumed that the count rate was caused by ²⁴²Pu entirely; if more normal isotopes were assumed, the Pu mass would be much larger.

Uranium Masses

Only one anomaly arose from the active-neutron measurements for uranium. The drum was already mentioned in the previous sections on the gamma-ray and passive-neutron measurements.

- The data for drum R1 indicates no uranium is present, strongly contrary to the declaration. The passive-neutron measurement shows no plutonium, whereas 317 g is declared and the gamma-ray spectrum has no indication of any fissile material. Either the declarations are wrong or the drum was not in the shuffler during these measurements. (After this result was obtained the drum was opened and in fact the contents did not include any uranium or plutonium.)