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TITLE RECENT EXPERIENCES OF SCRAP AND WASTE ASSAY USING NEUTRON COINCIDENCE COUNTING OF MATERIALS FROM F B-LINE AT THE SAVANNAH RIVER SITE

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RECENT EXPERIENCES OF SCRAP AND WASTE ASSAY USING NEUTRON COINCIDENCE COUNTING OF MATERIALS FROM F B-LINE AT THE SAVANNAH RIVER SITE

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ABSTRACT

The heterogeneous nature of scrap and waste poses unique problems for quantitative measurement. This is particularly true when the sample to be measured can have significant amounts of plutonium in addition to a variety of matrix components. One approach to addressing these difficulties is to combine gamma-ray and neutron counting techniques. A system combining a segmented gamma-ray scanner and neutron coincidence counter has recently been developed for the nondestructive assay of plutonium-bearing scrap and waste. Experience gained with the neutron coincidence counter portion of the system is described. Results obtained from the measurement of scrap and waste materials containing a few grams to a few hundred grams of plutonium are given. The effects of different matrices are evaluated and the role of special diagnostics is explored.

INTRODUCTION

Quantitative measurement of the plutonium in scrap and waste poses challenges, primarily because the samples are heterogeneous. This is particularly true when the sample has significant amounts of plutonium in addition to a variety of matrix components. Instruments that measure only gamma rays or neutrons can have large biases as a result of these sample characteristics. The primary cause of assay bias for gamma-ray techniques is attenuation, especially when the plutonium is in the form of lumps. Neutron measurements can suffer from varying α,n rates, self-shielding, and self-multiplication that arise from changing chemical composition and fissile mass of the sample. One approach to addressing these difficulties is combining gamma-ray and neutron counting techniques. As a general rule gamma-ray instruments perform optimally for lower density samples, whereas neutrons are more penetrating and can therefore be used to measure higher masses. As such the two methods are complementary and present the opportunity to make two independent measurements of the same sample. A system[1] combining a segmented gamma scanner (SGS) and neutron coincidence counter (NCC) has recently been developed to nondestructively assay plutonium-bearing scrap and waste from the F B-Line at the Savannah River Site. Samples that are measured on both the SGS and NCC yield results of high confidence when both instruments agree. When the instruments disagree, diagnostics are included that give guidance on what result is best for the particular sample being measured. This paper will focus on the experience gained with the NCC portion of the combined system. The results were obtained from measuring scrap and waste materials containing a few grams to a few hundred grams of plutonium. The effects of different matrices are evaluated and the role of special diagnostics is explored.

COUNTER AND SAMPLE DESCRIPTION

Figure 1 shows the combined SGS and NCC system.

SGS

The SGS consists of a collimated germanium detector and a sample rotation/translation stand. The sample is contained in a 5-gal. can and is moved vertically in discrete steps (segments) while rotating. Two measurements are made: a passive count in which the detector sees only gamma rays from the sample and a transmission count in which gamma rays from an external source are passed through the sample. The transmission measurement allows for the correction of sample self-attenuation by the matrix, while the evaluation of gamma-ray intensities from 100 to 400 keV corrects for attenuation from the plutonium particles themselves, the so-called lump correction.[2]

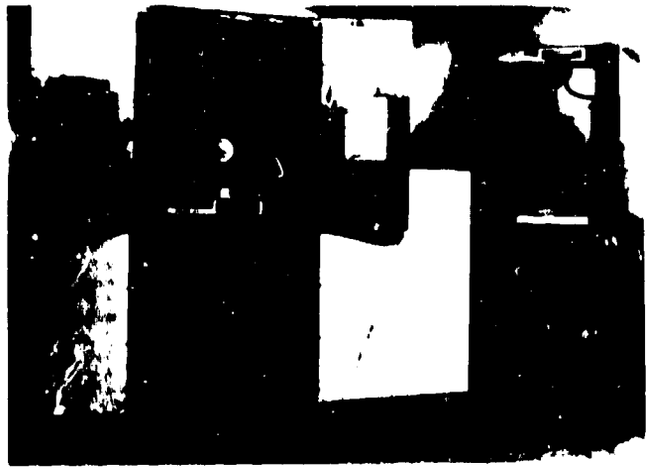

Fig. 1. Combined segmented gamma scanner (left) and neutron coincidence counter (right) for the Savannah River Site F B-Line.

NCC

As shown in Fig. 2, the NCC consists of a central cavity with a diameter slightly larger than the diameter of a 5-gal. can, surrounded by an annulus of 1-in. diam ^{3}He proportional detectors embedded in a high-density polyethylene moderator. Spontaneous fission neutrons emitted from the even isotopes of plutonium in the sample are counted using shift register electronics.[3] The total plutonium mass is calculated by combining the sample isotopics with the effective ^{240}Pu mass determined from measuring the time-correlated count rate (also called the coincidence or reals rate).

A sample is lowered into the central cavity by means of an elevator mechanism. Limit switches determine the location of the sample carrier and provide input to the software to correctly position the sample during the assay. Graphite endplugs increase the counter efficiency and flatten the axial response. Figure 3 shows the relative axial profile for both totals and reals measured with a ^{252}Cf source. The counter incorporates two rings of ^{3}He tubes. Thirty-six detector tubes compose the main ring providing the coincidence signal for the plutonium assay. A second, inner ring of three undermoderated tubes is sensitive to moderation effects in the sample. Figure 4 shows the tube layout, polyethylene shielding, and cadmium locations for the NCC. Cadmium lines the sample cavity preventing thermal neutrons from re-entering the sample and causing additional neutron multiplication. The outer ring of polyethylene and the cadmium layer between the inner and outer rings of polyethylene provides shielding from external sources allowing measurement of samples containing small amounts of plutonium. Table I summarizes data on NCC characteristics.

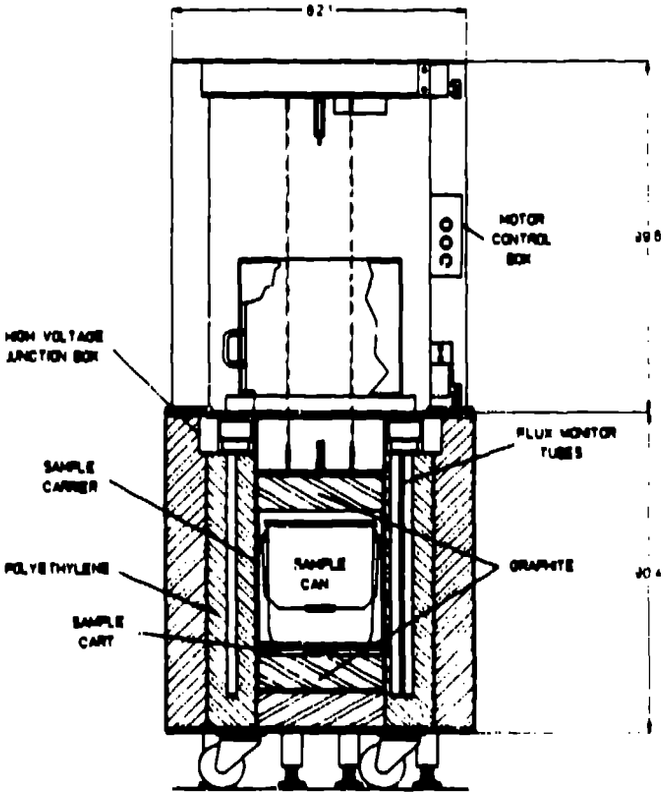

Fig. 2. Cross sectional view of the NCC showing the relation of sample to moderator and ^{3}He detectors.

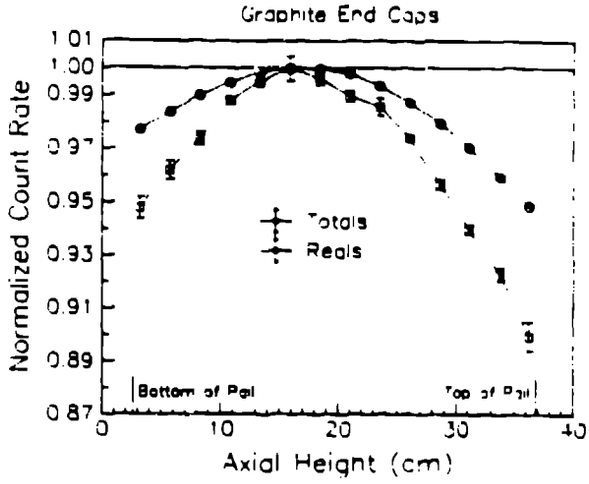

Fig. 3. Axial response profiles (totals and reals) for a ^{252}Cf point source.

The NCC software allows the user to control the data acquisition and analysis. Program options are divided into routine and supervisory levels. Passwords determine the level of access allowed. Routine operations consist of assay, background, and measurement control (bias and precision) activities. The supervisory options permit setting or changing such items as diagnostic values and calibration constants. The NCC currently has two calibrations that are used according to the sample type. One calibration is for low α,n samples, such as oxides and the other is used for high α,n samples, such as fluorides. In both cases, the calibrations are based on a quadratic relationship between the reals rate and the ^{240}Pu effective.[4] Figure 5 shows the two calibration curves that are discussed in the next section.

The assayed scrap comes from the mechanical line glove box in the F B-Line at Westinghouse Savannah River Site (SRS). The mechanical line handles mixed plutonium oxide and fluoride powder in preparation for the calciothermic reduction to plutonium metal. In the process of handling these powders, some are spilled on the floor of the process equipment or the glove box. These powders, called sweepings, may contain plutonium trifluoride, plutonium tetrafluoride, plutonium oxide, and calcium oxide. They are collected using a brush and scoop or hand-held vacuum cleaner, screened, and placed into a standard stainless steel container; the lid is sealed with tape. Each container is limited to 1000 g gross weight. The container is bagged from the glove box and stored in 5-gal. pails.

Sweepings are assayed to comply with the DOE 5633.3 requirement for a measured material balance. Results are used for material control and material accountability. Also, the throughput of the scrap recovery dissolver operation can be improved while maintaining nuclear safety conditions. In addition, storage space can be used more efficiently.

MATRIX EFFECTS AND DIAGNOSTICS

With two independent measurement instruments at the F B-Line facility, it is not necessary that both instruments always give the same assay result. If the assays from the two

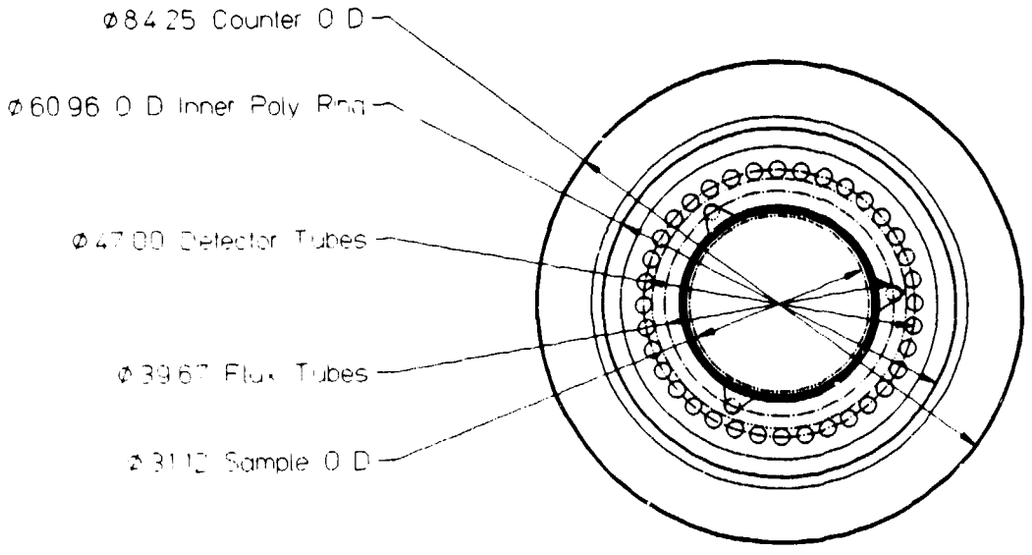

Fig. 4. Cross section of the NCC showing the tube layout.

Table I. Summary of Detector Characteristics	
Number of tubes	
main ring	36
flux monitor	3
Tube active length	24 in.
Pressure	4 atm.
Detector efficiency	23%
Die-away time	59 μs

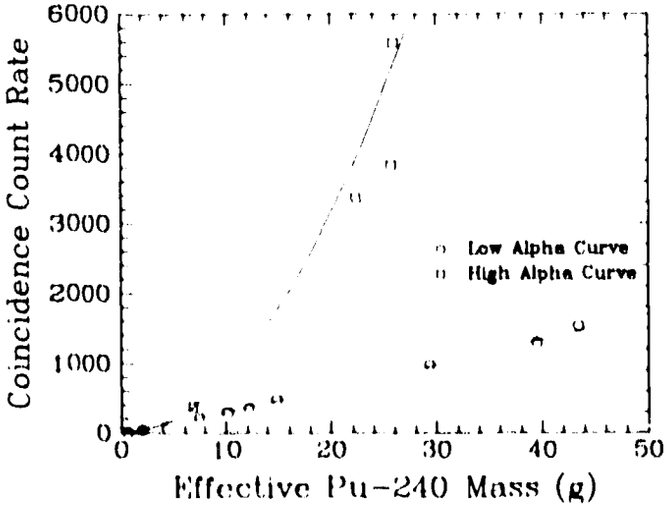

Fig. 5. Calibration curves for low and high α-n samples.

instruments agree, one can accept the value. Because of the complementary aspects of the SGS and the NCC, when the assay values disagree, one of the assay values is usually better than the other. For each of the instruments, diagnostic tools are incorporated into the analysis software to indicate the potential bias of the respective assays. If the assay values do not agree, then a decision must be made on the acceptability of either value. These diagnostic indicators guide the determination of the better value.

In NCC's, assays are performed by relating the measured coincidence count rate to the effective ^{240}Pu mass of the sample. In the ideal situation, the coincidence count rate is a linear function of the mass. In reality, however, the coincidence response is not linear because of multiplication in the sample. For well-characterized samples, techniques have been developed to adjust the measured coincidence count rate for self-multiplication.[5] The technique uses the ratio of the measured coincidence count rate to the measured total neutron count and the known plutonium isotopics of the sample to calculate a multiplication correction factor. The mass is then determined from the corrected coincidence count rate and a linear calibration curve. Multiplication corrections adjust for variations in sample geometry, density, and composition.

Without multiplication corrections, assays are based on calibration curves that include multiplication. These calibration curves are developed by using standards of known mass. The accuracy of the assays depends on how well characteristics of the samples being measured match the characteristics of the calibration standards. For waste and scrap materials, we assume that very little information is available concerning the geometry or the composition of the sample being measured and multiplication corrections are generally not possible. As a result, F B-Line NCC assays use nonmultiplication-corrected calibration curves.

Moderating materials and materials with high (α,n) yields in the samples cause problems in NCC assays. Waste can contain a variety of materials from the process line.

Sometimes the materials contain high levels of hydrogen that acts as a neutron moderator within the sample. Moderator, generally in the form of moisture or an abundance of plastic materials such as gloves and containers, softens the neutron spectra. Spectral changes impact multiplication in the sample and affect the overall detector efficiency. Overall the softer spectrum alters the coincidence count rate relative to the effective ^{240}Pu mass. This affects the applicability of the calibration curve.

One of the calibration curves for the F B-Line NCC is made with well-known PuO_2 standards. Oxide samples have low (α,n) yields. If the sample materials come from parts of the process line where the plutonium chemical form is a low (α,n) emitter, this calibration curve is adequate. However, in parts of the process line, the plutonium chemistry is different and there are other low-Z materials, such as fluorides, present which have much higher (α,n) yields compared to oxides. High (α,n) yields significantly increase the coincidence rate per effective ^{240}Pu mass within the sample. The oxide calibration curve is not applicable for these materials. Most of the assay scrap material is of a high (α,n) nature and therefore the NCC has a separate calibration curve for these samples.

Two key indicators are used in the NCC software to determine the presence of significant matrix effects. Increased moderation in the sample can be detected by using the flux monitor signal. A softer spectrum emerging from the sample increases the flux monitor response relative to the main ring signal. The ratio of the total count rate in the main ring to the total count rate in the flux monitor decreases as moderation increases. When the T/T_{flux} ratio decreases below a preset value, a diagnostic message is printed. This indicates that the NCC value is questionable and that the SGS value should probably be accepted. The NCC value would generally underpredict the effective ^{240}Pu mass for the case of increased moderation. In the analysis, no adjustments are available to the NCC data when the T/T_{flux} diagnostic is indicated.

The ratio of the coincidence count rate to the total count rate (R/T) in the ring of tubes is a second diagnostic and it is used to determine if high (α,n) materials are present. When very high (α,n) backgrounds are present, the R/T ratio decreases significantly. The resulting coincidence count rate also increases requiring a separate calibration curve.

A lower R/T ratio for the sample indicates the presence of a high (α,n) source. For the F B-Line application, we characterized the kinds of materials that would occur in the process line. From this information, we established two calibration curves; a low (α,n) curve and a high (α,n) curve. The oxide curve is the low (α,n) curve. If the operator does not know the process origin of the sample being measured, the R/T flag can be used to determine that the high (α,n) curve should be used for the assay. Eventually, software may be modified to automatically use the high (α,n) curve when the R/T flag triggers.

The high and low (α,n) calibration curves are shown in Fig. 5. For the same coincidence count rate, two very different values for the effective ^{240}Pu mass are obtained from the two curves. Data for both curves are fitted to second-order polynomials that pass through the origin. The fit coefficients are given in Table II. The low (α,n) mass curve is well characterized from known PuO_2 standards. The high (α,n) curve is developed from typical process line samples. These samples are not as well characterized as the PuO_2 standards. Thus the high (α,n) curve does not fit the data as well. However, the curve represents the available calibration data, and it is adequate for the high (α,n) F B-Line scrap samples.

Table II. Calibration Curve Coefficients
Curve: $R = a_0 + a_1 * m + a_2 * m^2$

	Low (a,n) Curve	High (a,n) Curve
a_0	0.00	0.00
a_1	30.27	3.14
a_2	0.08	7.76

m = mass of ^{240}Pu effective

RECENT EXPERIENCE AT SRS

The SGS and NCC instruments were used to study sweeping samples from the F B-Line at SRS. Following are discussions of the measurements on the sweeping samples.

A typical day's operation is shown in Table III. Backgrounds are run on both SGS and NCC instruments. On the NCC, the measurement control-bias measurements are made with a reference californium source. The decay-corrected expected coincidence rate is divided by the measured coincidence rate to give a normalization constant (K). If the new value of K differs from the old value by less than three standard deviations of the new value, then the old value is retained. The measurement control standard for the SGS is a plutonium oxide waste standard, B-88. Figure 6 shows the SGS assay on B-88. The results show that the average SGS result agrees with the reference value. Runs of the waste standard B-88 on the NCC are shown in Fig. 7. The oxide calibration curve was used. The average NCC assay is 10% low. This sample is known to contain a significant amount of plastic. The T/T_{flux} diagnostic message is generated and the NCC would generally under predict the effective ^{240}Pu mass for the case of increased moderation.

As part of the measurement control program, we run a sweeping standard on each instrument before measuring samples. The working standards are actual sweepings taken from various locations in the F B-Line mechanical glove box. They were packaged for characterization by calorimetry and gamma isotopic measurements. The uncertainty associated with the calorimeter and gamma isotopic instruments has been determined to be between 2% and 7%. Four working standards were used to establish the high (α,n) fluoride calibration curve for assay of the sweeping standards on the NCC. Figure 8, which shows Standard 4361 assay results on the SGS, indicates that the average SGS value agrees with the reference value. Figure 9 shows the assay results of Standard 4361 run on the NCC. The results show that the NCC assay on the average is 4% high. This is not unexpected because the sweepings are known to contain fluoride that yields high (α,n).

The other working standard is Standard 4362. Figure 10 is a measurement control chart showing the assay results of the standard run on the SGS. Agreement is quite good indicating the new lump correction software is effective, and this SGS remained in control during the time period over which the sweepings were measured. Results of the Standard 4362 run on the NCC are shown in Fig. 11. They show that the NCC assay on the average is higher than the reference by about 6%. This bias is due to the systematic error in the calibration curve as generated with the four fluoride standards.

Table III. Example of Daily Operations

Number	Date	Time	Counter	Sample Type	Result	Comments
1	21-May	10:20	NCC	Bkgd	T = 988 ± 1.3; R = 0.03 ± 0.45; FM = 13.2 ± 0.15	
2	21-May	10:54	SGS	Bkgd		Resolution too good
3	21-May	10:49	NCC	Normalization		Normalization constant retained
4	21-May	11:00	SGS	Standard B-88	3.50 vs 3.58 reference value	Bias test passed
5	21-May	12:41	NCC	Standard 4362	456 ± 32 (OK)	
6	21-May	12:43	SGS	Standard 4361	110 ± 1 (OK)	
7	21-May	13:07	NCC	Can 1	248 ± 21 (C)	R/T = 0.0056; test failed
8	21-May	13:08	SGS	Can 2	241 ± 9	
9	21-May	13:27	NCC	Can 2	240 ± 18 (C)	R/T = 0.0085; test failed
10	21-May	13:29	SGS	Can 1	268 ± 7	
11	21-May	13:48	NCC	Can 1	276 ± 20 (C)	R/T = 0.00070; test failed
12	21-May	13:49	SGS	Can 2	237 ± 8	
13	21-May	14:08	NCC	Can 2	235 ± 18 (C)	R/T = 0.0081; test failed
14	21-May	14:09	SGS	Can 1	269 ± 4	
15	21-May	15:20	NCC	Standard ^{252}Cf	Chi square = 0.53	Precision OK

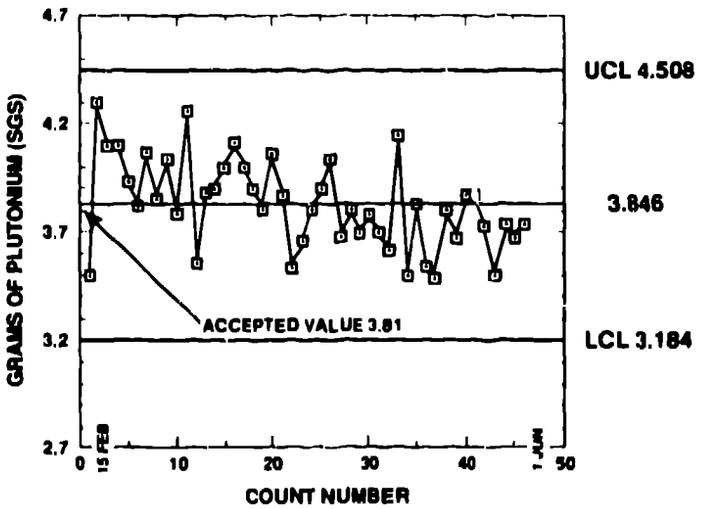

Fig. 6. Historical SGS results of PuO_2 Standard B-88 showing accepted value, average measured result, and confidence limits.

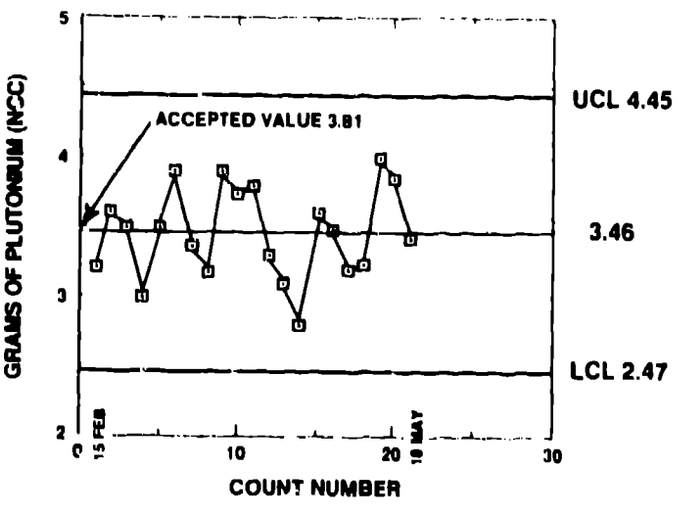

Fig. 7. Historical NCC results of PuO_2 Standard B-88 showing accepted value, average measured result, and confidence limits.

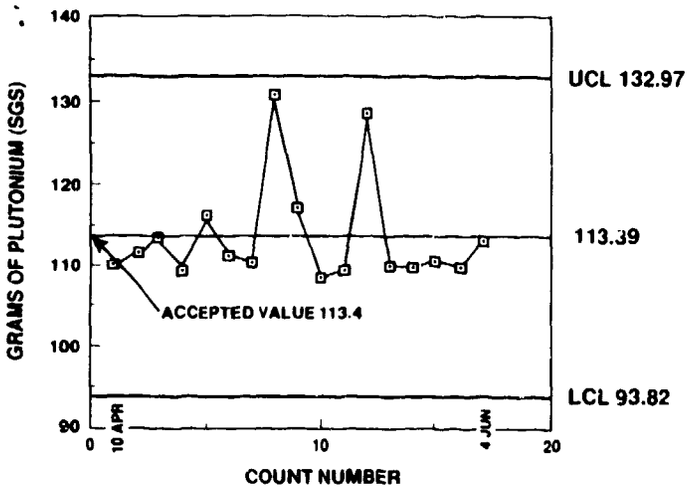

Fig. 8. SGS results of sweepings Standard 4361 from April to June 1990 showing accepted value, average measured result, and confidence limits.

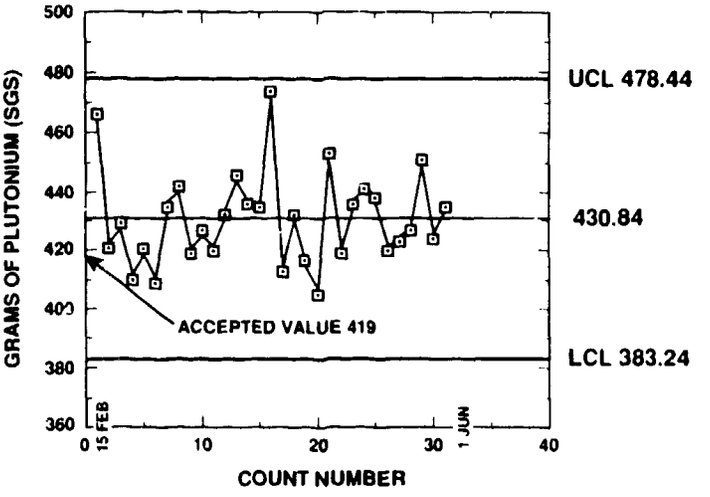

Fig. 10. Results of SGS measurement of sweepings Standard 4362 from February to June 1990, showing accepted value, average measured result, and confidence limits.

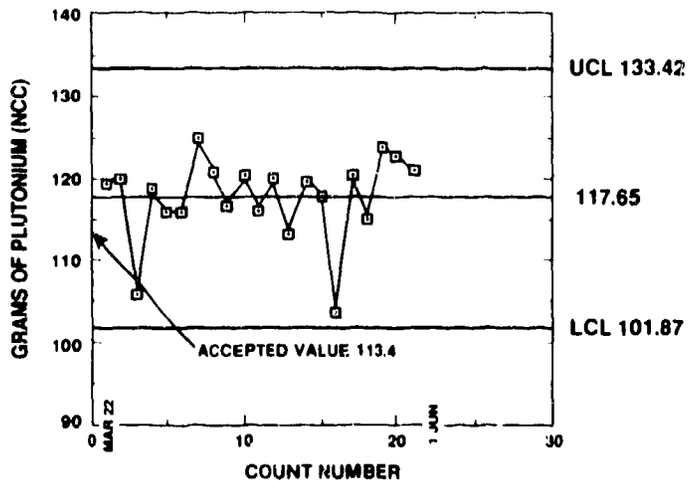

Fig. 9. NCC results of sweepings Standard 4361 from April to June 1990, showing accepted value, average measured result, and confidence limits.

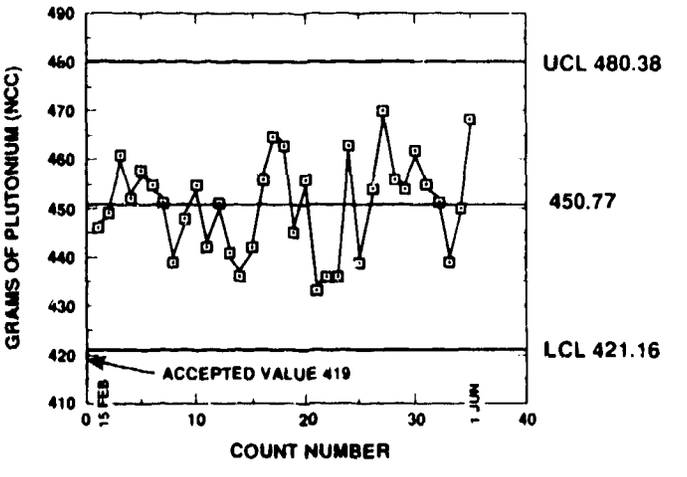

Fig. 11. Results of NCC measurement of sweepings Standard 4362 from February to June 1990, showing accepted value, average measured result, and confidence limits.

Assay results of approximately 100 sweeping samples run on the SGS and NCC are shown in Fig. 12. Results show that the SGS and NCC generally agree within the measurement uncertainties. The R/T ratio on the NCC results is usually flagged indicating the presence of high (α,n). This diagnostic is useful because it verifies the presence of high fluorides in the sample and the fluoride calibration curve is the correct curve to use. Because the NCC and SGS results agree, the NCC calibration curve appears to be adequate for the high (α,n) F B-Line sweeping samples. Figure 13 further defines the low mass sweepings run on the SGS and NCC.

SUMMARY

Experience of assaying sweeping samples at SRS shows that the NCC and SCS can perform assays with no significant bias on most F B-Line sweeping samples. The SGS and NCC results agree within the uncertainties, and we are confident that we have a reliable assay result and a reliable error estimate.

We are now able to meet the DOE requirement of a measured material balance in the F B-Line. Also, process recovery dissolver throughput can be improved and nuclear safety conditions can be maintained. Results can be used for material control and material accountability. In addition, vault storage space can be used more efficiently.

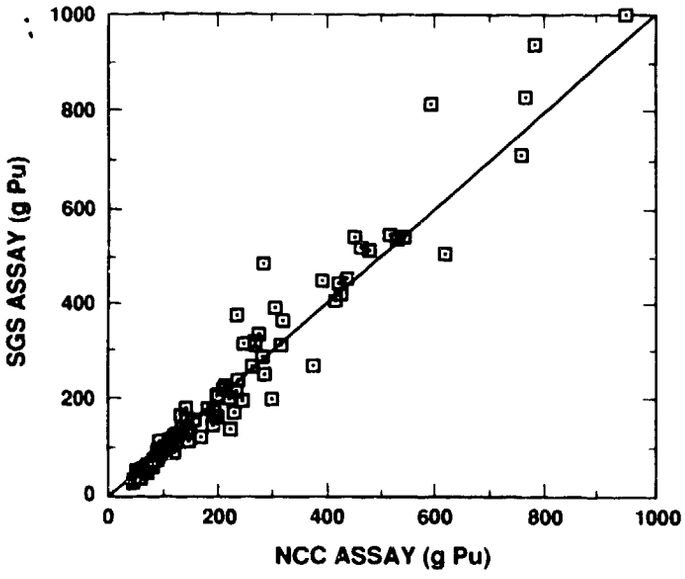

Fig. 12. Comparison of NCC and SGS assays for sweepings samples containing a few grams to a few hundred grams of plutonium.

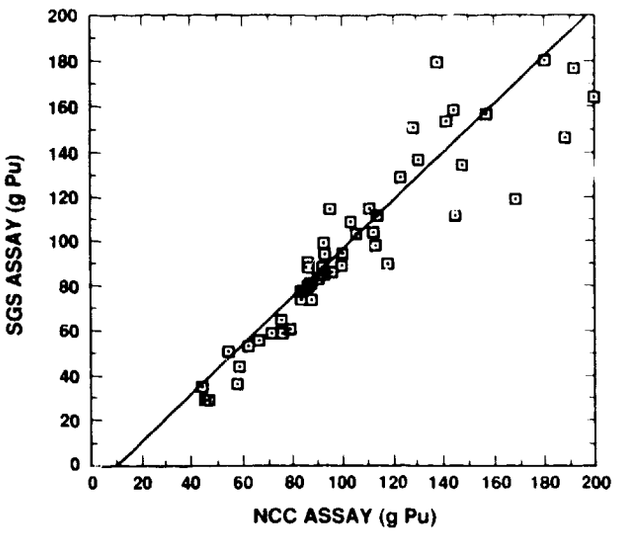

Fig. 13. Comparison of NCC and SGS assay results of sweepings samples containing up to 200 g of plutonium.

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