Designation: C 1500 – 02

Standard Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting[1]

This standard is issued under the fixed designation C 1500; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This test method describes the nondestructive assay of plutonium in forms such as metal, oxide, scrap, residue, or waste using passive neutron multiplicity counting. This test method provides rapid results that are usually more accurate than conventional neutron coincidence counting. The method can be applied to a large variety of plutonium items in various geometries in cans, 208-L drums, or 1900-L Standard Waste Boxes. It has been used to assay items whose plutonium content ranges from 1 g to 1000's of g.

1.2 There are several electronics or mathematical approaches available for multiplicity analysis, including the shift register, the Euratom Time Correlation Analyzer, and the List Mode Module, as described briefly in Ref. **(1)**.[2]

1.3 This test method is primarily intended to address the assay of ^{240}Pu-effective by moments-based multiplicity analysis using shift register electronics **(1, 2)** and high efficiency neutron counters specifically designed for multiplicity analysis. This test method requires knowledge of the relative abundances of the plutonium isotopes to determine the total plutonium mass.

1.4 This test method may also be applied to modified neutron coincidence counters which were not specifically designed as multiplicity counters, with a corresponding degradation of results.

2. Referenced Documents

2.1 *ASTM Standards:*

C 859 Terminology Relating to Nuclear Materials[3]

C 1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectroscopy[3]

C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting[3]

C 1458 Test Method for Nondestructive Assay of Plutonium, Tritium, and ^{241}Am by Calorimetric Assay[3]

3. Terminology

3.1 Terms shall be defined in accordance with Terminology C 859 except for the following:

3.2 *alpha (α), n*—the ratio of the uncorrelated neutron emission rate from (α,n) reactions to the spontaneous neutron emission rate from a non-multiplying sample (see Ref. **(1)** for equation).

3.3 *coincidence gate length (G), n*—the time interval following the detection of a neutron during which additional neutron counts are considered to be in coincidence with the original neutron. In Fig. 1, this is the length of time the ($R + A$) and (A) gates are set to accept neutron counts.

3.3.1 *gate fractions, n*—the fraction of the total coincidence events that occur within the coincidence gate.

3.3.2 *doubles gate fraction* (f_d), *n*—the fraction of the theoretical double coincidences that can be detected within the coincidence gate (see Eq 1).

3.3.3 *triples gate fraction* (f_t), *n*—the fraction of the theoretical triple coincidences that can be detected within the coincidence gate (see Eq 2).

3.4 *die-away time (τ), n*—the average mean life-time of the neutron population as measured from the time of emission to the time of detection, escape, or absorption. Die-away time is a function of the counter assembly design and the assay item. Fig. 1 illustrates the decreasing probability of detection as a function of time.

3.5 *doubles (D), n*—the doubles are equivalent to the reals rate and represents the number of double neutron coincidences/s. The doubles may be determined from the coincidence shift register directly or by reduction of the multiplicity ($R + A$) and (A) histograms **(1)**.

3.6 *efficiency (ε), n*—this is usually taken to be the absolute neutron detection efficiency, which is calculated from the ratio of the measured neutron count rate to the declared neutron emission rate of a non-multiplying reference source.

3.7 *factorial moment, n*—this is a derived quantity representing a summation of the neutron multiplicity distribution weighted by certain factors (see Ref. **(1)** for equation).

3.8 *item, n*—the entire container being measured and its contents.

3.9 *multiplicity distribution, n*—this is the distribution of the number of neutrons emitted in a fission event. This number can vary from 0 to 5 or more.

[1] This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.10 on Non Destructive Assay.

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[2] The boldface numbers in parentheses refer to the list of references at the end of this standard.

[3] *Annual Book of ASTM Standards*, Vol 12.01.

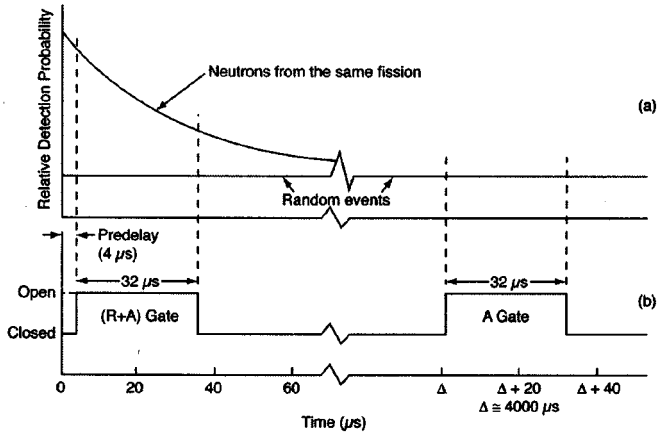

FIG. 1

(a) Simplified porbability distribution showing the approximately exponential decay, as a function of time, for detecting a second neutron from a single fission event. The probability of detecting a random neutron is constant with time. (b) Typical coincidence timing parameters.

3.9.1 *spontaneous fission neutron multiplicities* (ν_{s1}, ν_{s2}, ν_{s3}), *n*—the factorial moments of the spontaneous fission neutron multiplicity distribution. For the multiplicity analysis of Pu materials the spontaneous fission nuclear data for ^{240}Pu is used to calculate these moments **(3)**. One commonly used set of moments is ν_{s1} = 2.154, ν_{s2} = 3.789, ν_{s3} = 5.211 **(23)**.

3.9.2 *induced fission neutron multiplicities* (ν_{i1}, ν_{i2}, ν_{i3}), *n*—the factorial moments of the induced fission neutron multiplicity distribution. Typically multiplicity analysis will utilize the data from fast neutron-induced fission of ^{239}Pu to calculate these moments **(3)**. One commonly used set of moments is ν_{i1} = 3.163, ν_{i2} = 8.240, ν_{i3} = 17.321 **(23)**.

3.10 *point model*, *n*—the mathematical model used to analyze multiplicity counting data. The model assumes that the neutron detector efficiency and the probability of fission are constant across the item, as though it were a point source.

3.11 *shift-register-based coincidence circuit*, *n*—an electronic circuit for determining totals *T*, reals plus accidentals (*R* + *A*), and accidentals (*A*) in a selected count time *t* **(4, 5)**. The terminology used in this test method refers specifically to shift-register electronics. Fig. 1 shows the probability of detecting a neutron as a function of time and illustrates the time intervals discussed.

3.11.1 *totals*, *n*—the total number of neutrons detected during the count time.

3.11.2 *reals plus accidentals, (R + A), n*—the number of neutrons detected in the (*R* + *A*) gate period (Fig. 1) following the initial detection of each neutron **(4)**. These events are due to neutrons that are coincident with the given neutron (reals) and to neutrons that are not correlated with the given neutron (accidentals). This is a measured quantity.

3.11.3 *accidentals (A), n*—the number of neutrons detected in the (*A*) gate period (Fig. 1) following the initial detection of each neutron **(4)**. These neutrons are not correlated with the initial neutron. They come from many different sources and their count rate is assumed to be constant from the item being assayed. This quantity is measured by interrogating the (*A*) gate time interval window that occurs long after the expected lifetime of coincident neutrons in the counting chamber. This is a measured quantity.

3.11.4 *reals (R), n*—the number of coincident neutrons detected in (*R* + *A*) gate intervals immediately following the detection of each neutron during the count time **(4)**. This quantity is calculated from the measured (*R* + *A*) and (*A*) quantities.

3.11.5 *neutron counting multiplicity, n*—the number of neutrons within the coincidence gate for each trigger event in the shift register.

3.12 *net neutron leakage multiplication (M), n*—the ratio of the net number of neutrons leaving the item to the number initially produced by spontaneous fission and (α,n) reactions **(6)**.

3.13 *passive mode*, *n*—determines the total spontaneous fissioning mass in the measured item through the detection of emitted neutrons rather than neutrons from fissions induced by external interrogation sources.

3.14 *pre-delay*, *n*—the coincidence circuit has a pre-delay immediately after a neutron has been detected to allow the amplifiers to recover and prepare to detect subsequent neutrons **(4)**. This principle is shown in Fig. 1.

3.15 *singles (S), n*—the singles are equivalent to the totals/s representing the total neutron detection rate.

3.16 *triples (T), n*—The triple neutron coincidence rate is a derived quantity obtained from the factorial moments of the multiplicity (*R* + *A*) and (*A*) histograms **(1)**. It may be visualized as the count rate for three neutrons in coincidence.

4. Summary of Test Method

4.1 The item is placed in the sample chamber or "well" of the multiplicity counter, and the emitted neutrons are detected by the ^{3}He tubes that surround the well.

4.2 The detected neutron multiplicity distribution is processed by the shift register electronics package to obtain the number of neutrons of each multiplicity in the (*R* + *A*) and (*A*) gates.

4.3 The first three moments of the (*R* + *A*) and (*A*)

multiplicity distributions are computed to obtain the singles (or totals), the doubles (or reals), and the triples. Using these three calculated values, it is possible to solve for 3 unknown item properties, the ^{240}Pu-effective mass, the self-multiplication, and the α ratio. Details of the calculations may be found in Annex A1.

4.4 The total plutonium mass is then determined from the known plutonium isotopic ratios and the ^{240}Pu -effective mass.

4.5 Corrections are routinely made for neutron background, cosmic ray effects, small changes in detector efficiency with time, and electronic deadtimes.

4.6 Optional algorithms are available to correct for the biases caused by spatial variations in self-multiplication or changes in the neutron die-away time.

4.7 Multiplicity counters are carefully designed by Monte Carlo techniques to minimize variations in detection efficiency caused by spatial effects and energy spectrum effects. Corrections are not routinely made for neutron detection efficiency variations across the item, energy spectrum effects on detection efficiency, or neutron capture in the item.

5. Significance and Use

5.1 This test method is useful for determining the plutonium content of items such as impure Pu oxide, mixed Pu/U oxide, oxidized Pu metal, Pu scrap and waste, Pu process residues, and weapons components.

5.2 Measurements made with this test method may be suitable for safeguards or waste characterization requirements such as:

5.2.1 Nuclear materials accountability,

5.2.2 Inventory verification **(7)**,

5.2.3 Confirmation of nuclear materials content **(8)**,

5.2.4 Resolution of shipper/receiver differences **(9)**,

5.2.5 Excess weapons materials inspections **(10, 11)**,

5.2.6 Safeguards termination on waste **(12, 13)**,

5.2.7 Determination of fissile equivalent content **(14)**.

5.3 A significant feature of neutron multiplicity counting is its ability to capture more information than neutron coincidence counting because of the availability of a third measured parameter, leading to reduced measurement bias for most material categories. This feature also makes it possible to assay some in-plant materials that are not amenable to conventional coincidence counting, including moist or impure plutonium oxide, oxidized metal, and some categories of scrap, waste, and residues **(10)**.

5.4 Calibration for many material types does not require representative standards. Thus, the technique can be used for inventory verification without calibration standards **(7)**, although measurement bias may be lower if representative standards were available.

5.4.1 The repeatability of the measurement results due to counting statistics is related to the quantity of nuclear material, the (α,n) reaction rate, and the count time of the measurement **(15)**.

5.4.2 For certain materials such as small Pu items of less than 1 g, some Pu-bearing waste, or very impure Pu process residues where the (α,n) reaction rate overwhelms the triples signal, multiplicity information may not be useful because of the poor counting statistics of the triple coincidences within practical counting times **(12)**.

5.5 For pure Pu metal, pure oxide, or other well-characterized materials, the additional multiplicity information is not needed, and conventional coincidence counting will provide better repeatability because triple coincidences are not used. Conventional coincidence information can be obtained either by changing to a coincidence counter, or analyzing the multiplicity data in coincidence mode.

5.6 The mathematical analysis of neutron multiplicity data is based on several assumptions that are detailed in Annex A1. The most important is the assumption that the item is a point in space, so that neutron detection efficiency, die-away time, and multiplication are constant across the entire item **(16, 17)**.

5.6.1 Bias in passive neutron multiplicity measurements is related to deviations from the “point model” such as variations in detection efficiency, matrix composition, or distribution of nuclear material in the item’s interior.

5.6.2 Heterogeneity in the distribution of nuclear material, neutron moderators, and neutron absorbers may introduce biases that affect the accuracy of the results. Measurements made on items with homogeneous contents will be more accurate than those made on items with inhomogeneous contents.

6. Interferences

6.1 For measurements of items containing several hundred grams of plutonium metal or more, multiplication effects are not adequately corrected by this method **(18)**. A variable-multiplication bias correction is required.

6.2 For items with high (α,n) reaction rates, the additional uncorrelated neutrons will significantly increase the accidental coincidence rate. The practical application of multiplicity counting is usually limited to items where the ratio of (α,n) to spontaneous fission neutrons is about 7 **(7)**.

6.3 For measurement of large items with high (α,n) reaction rates, the neutrons from (α,n) reactions can introduce biases if their energy spectra are different from the spontaneous fission energy spectrum. The ratio of the singles in the inner and outer rings can provide a warning flag for this effect **(19)**.

6.4 Neutron moderation by low atomic mass materials in the item affects neutron detection efficiency, neutron multiplication in the item, and neutron absorption by poisons. For moderate levels of neutron moderation, the multiplicity analysis will automatically correct the assay for changes in multiplication. A correction for capture in neutron poisons or other absorbers is not available, so that a bias can result in measurements of such items.

6.5 It is important to keep neutron background levels from external sources as low and constant as practical for measurement of low Pu mass items. High backgrounds may produce a bias, depending on the item’s mass and self-multiplication.

6.6 Cosmic rays can produce single, double, and triple neutrons from spallation events within the detector or nearby hardware. The relative effect is greatest on the triples, and next greatest on the doubles. Cosmic ray effects become significant for assay items containing large quantities of high atomic number matrix constituents and small gram quantities of plutonium. Multiplicity data analysis software packages should

include correction algorithms for count bursts caused by cosmic rays.

6.7 Other spontaneous fission nuclides (for example, curium or californium) will increase the coincident neutron count rates, causing a positive bias in the plutonium assay that multiplicity counting does not correct for. The triples/doubles ratio can sometimes be used as a warning flag.

7. Apparatus

7.1 *Multiplicity Counters*:

7.1.1 Neutron multiplicity counters are similar in design and construction to conventional neutron coincidence counters, as described in Test Method C 1207. Both are thermal neutron detector systems that utilize polyethylene-moderated ^{3}He proportional counters. However, multiplicity counters are designed to maximize neutron counting efficiency and minimize neutron die-away time, with detection efficiencies that are much less dependent on neutron energy. Multiplicity counters have 3 to 5 rings of ^{3}He tubes and absolute neutron detection efficiencies of 40 to 60 %, whereas conventional coincidence counters have 1 or 2 rings of ^{3}He tubes and efficiencies of 15 to 25 %. A multiplicity counter for the assay of cans of plutonium is illustrated in Fig. 2 **(20)**.

7.1.2 Multiplicity counters are designed to keep the radial and axial efficiency profile of the sample cavity as flat as possible (within several percent) to minimize the effects of item placement or item size in the cavity. Provision for reproducible sample positioning in the cavity is still recommended for best accuracy.

7.1.3 Multiplicity counters are designed with a nearly flat neutron detection efficiency as a function of the neutron energy spectrum, largely through the use of multiple rings of ^{3}He tubes placed at different depths in the polyethylene moderator material.

7.1.4 Multiplicity counters usually have a thick external layer of polyethylene shielding to reduce the contribution of background neutrons from external sources.

7.1.5 Existing conventional neutron coincidence counters are sometimes used for multiplicity analysis. The quality of the multiplicity results will depend on the extent to which the converted counters meet the multiplicity design criteria given above.

7.2 *Multiplicity Electronics*:

7.2.1 An example of the physical layout of the ^{3}He tubes and amplifier electronics on a multiplicity counter is illustrated in Fig. 2. The junction box usually contains 20 or more fast preamp/discriminator circuits to allow operation at very high count rates with short multiplicity electronic deadtimes. The ^{3}He tubes require a high voltage power supply, and the electronics require a +5 volt DC power supply. Depending on the multiplicity electronics package being used, it may be necessary to provide separate +5 V or HV power supplies.

7.2.2 Some multiplicity junction boxes include a derandomizer circuit that holds pulses that are waiting to enter the shift register, thus eliminating input synchronization losses **(21)**. With a derandomizer circuit, a conventional shift register can be operated at count rates approaching 2 MHz with virtually no synchronizer counting losses.

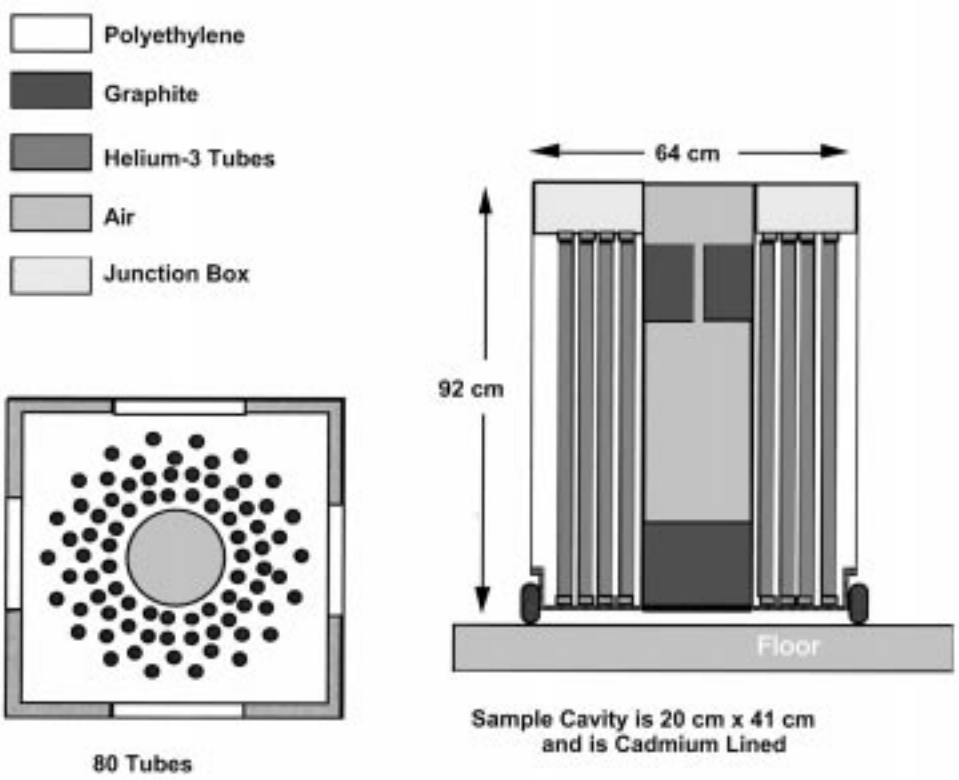

FIG. 2 Design Schematic for a Plutonium Multiplicity Counter. In this cross section of the counter, 80 ^{3}He tubes are arranged around the sample cavity. The space between the tubes is filled with polyethylene, and graphite above and below the sample cavity scatters and reflects neutrons. The junction box contains the fast preamp/discriminators.

7.2.3 A predelay circuit is usually included at the input to the multiplicity shift register to reduce the effect of small electronic deadtimes or pulse pileup effects in the ^{3}He tubes and eliminate a counting imbalance or "bias" between the *R+A* and *A* multiplicity distributions **(4)**.

7.2.4 A multiplicity shift register is required to measure the neutron multiplicity distributions in the *R+A* and *A* coincidence gates **(5)**. This electronics provides the same data as a conventional shift-register, and in addition records the number of times each multiplicity occurs in the *R+A* and *A* coincidence gates.

7.2.5 Software packages are needed to acquire and analyze data from the multiplicity shift register. Measurement control options, quality control tests, and calibration and least-squares fitting options are also needed in the software.

8. Hazards

8.1 *Safety Hazards*—Consult qualified professionals as needed.

8.1.1 It is recommended that a criticality safety evaluation be carried out if fissile material is to be measured, especially before assay of unknown items. The measurement chamber approximates a reflecting geometry for fast neutrons.

8.1.2 Precautions should be taken to avoid contact with high voltage. The ^{3}He tubes require low current high voltage power supplies.

8.1.3 Precautions should be taken to prevent inhalation, ingestion, or spread of plutonium contamination during item handling operations. All containers should be surveyed on a regular basis with an appropriate monitoring device to verify their continued integrity.

8.1.4 Precautions should be taken to minimize personnel exposure to radiation.

8.1.5 Counting chambers may contain a cadmium liner. Precautions should be taken to prevent the inhalation or ingestion of cadmium. It is a heavy metal poison. Cadmium shielding should be covered with nontoxic materials.

8.1.6 Pinch point and lifting hazards may be present during the loading and unloading of heavy items with multiplicity counters. Mechanical aids, such as a hoist, should be used for movement of heavy items.

8.1.7 The weight of the instrument may exceed facility floor loading capacities. Check for adequate floor loading capacity before installation.

8.2 *Technical Hazards*:

8.2.1 High mass, high α items will produce large count rates with large accidental coincidence rates. Very long count times may be required to obtain an assay result, or sometimes it is not possible to get a meaningful result.

8.2.2 Total counting rates should be limited to about 900 kHz to limit the triples deadtime correction to about 50 % and to ensure that less than 25 % of the shift register steps are occupied. Otherwise incorrect assay results may be obtained due to inadequate electronic deadtime corrections.

8.2.3 High gamma-ray exposure levels from the sample may interfere with the neutron measurement through pile-up effects if the dose is higher than 1 R/h at the ^{3}He tubes unless counter design takes gamma-ray exposure levels into account.

9. Preparation of Instruments

9.1 Perform initial multiplicity counter setup.

9.1.1 It is recommended that the counter be set up and used in an area with a range of temperature and humidity typical of an air-conditioned office environment, although newer electronics packages are specified to operate over the range of 0 to 50°C, and 0 to 95 % humidity. Movement of radioactive material in the vicinity of the counter should be avoided while measurements are in progress if the background count rates can change by 10 % or more.

9.1.2 Set up the initial detector, data collection, and data analysis parameters in the software code as recommended by the supplier. Turn on the quality-control tests in the analysis code, as described in Section 11.

9.1.3 For all measurements, split up the available count time into a series of many smaller runs.

9.2 Perform detector characterization measurements. These initial measurements will provide some of the initial detector parameters needed for setup.

9.2.1 Measure the room background singles, doubles, and triples rates to make sure that they are reasonable and no ^{3}He detector breakdown is indicated. These count rates can be used as initial measurement control values. Typical singles, doubles, and triples count rates are 100 to 1000 cps, 1 to 2 cps, and 0.1 to 0.2 cps, resp.

9.2.2 Perform an initial neutron source measurement to provide a reference value that can be used for measurement control purposes. This can be done with a ^{252}Cf reference source that will be readily available in the future, or with a physical standard that is not likely to change its shape, density or chemical form. If a ^{252}Cf source is used, the ^{250}Cf content should be low enough to allow decay corrections using the known half-life of ^{252}Cf alone. The source or standard should be placed in a reproducible location within the normal assay volume of the measurement chamber.

9.2.3 Using the reference source of known neutron yield, determine the neutron detection efficiency ϵ of the multiplicity counter (See Ref. **(1)** for equations). The isotopic data and neutron yield for the ^{252}Cf source should be certified to a national standard. The neutron singles rate should be corrected for background, electronic deadtime, and source decay. This is an excellent diagnostic that tests the ^{3}He detectors, the fast preamp/discriminator electronics chain, all hardware and software configurations, the counter's design specifications, and any effect of the detector's surroundings. The detection efficiency is also used later as part of the calibration process.

9.2.4 Verify that the detector die-away time τ is as expected from the manufacturer or from Monte Carlo calculations by re-measuring the ^{252}Cf reference source at a different gate length that differs by a factor of 2 (See Ref. **(1)** for equations). Some multiplicity counters will have more than one significant component to their die-away curves, so this calculation may yield somewhat different die-away times with different choices of gate length. The most appropriate choice of gate lengths for this test are those that bracket the expected die-away time.

9.2.5 Verify that the coincidence gate width *G* is set close to 1.27τ to obtain the minimum relative error for the assay **(22)**. At high count rates, it may be necessary to set the gate width

to a smaller value to keep the highest observed multiplicities in the (*R* + *A*) and (*A*) distributions under 128 to minimize the multiplicity deadtime correction **(23, 24, 25)**.

9.2.6 It is strongly recommended that the coincidence and multiplicity deadtime coefficients be checked if feasible because multiplicity data analysis requires careful deadtime corrections for the singles, doubles, and triples count rates. Ref. **(1)** provides an example of typical deadtime correction equations and a common procedure for determining them. For multiplicity counters, typical values for the doubles deadtime coefficient are in the range of 0.1 to 0.6 µs, and typical values for the triples deadtime coefficient are in the range of 25 to 170 ns.

9.2.7 A series of 40 or more precision runs with the same item left in the counter can be carried out. This will provide some indication of the run-to-run stability of the electronics, and check that the statistical error propagation is being done correctly.

10. Calibration

10.1 Because multiplicity counters are used to assay or verify a wide variety of impure plutonium items, representative physical standards are usually not available, and it is possible to calibrate the counter without them. Instead, the singles, doubles, and triples equations are solved directly for multiplication *M*, α, and effective ^{240}Pu mass m_{eff} using a series of measured detector parameters **(1)**. The solution will provide an accurate assay to the extent that the plutonium items satisfy the assumptions used in multiplicity analysis, as described in Annex A1.

10.2 Adjust the detection efficiency ε for the difference in efficiency between californium and plutonium by Monte Carlo calculations or by measurement of a non-multiplying representative standard. The magnitude of the adjustment will depend on the actual multiplicity detector being used, but will typically be in the range of 1 to 2 %. If no other information is available, set the plutonium detection efficiency to be 1.02 times the californium detection efficiency.

10.3 Determine the actual fraction of the doubles that are counted within the gate width *G*. The doubles gate fraction f_d is calculated from the singles and doubles rates measured with a ^{252}Cf reference source (the parameters are defined in Section 3):

$$f_d = \frac{2\nu_{s1}D}{\epsilon\nu_{s2}S} \quad (1)$$

10.4 Determine a preliminary value for the fraction of the triples that are counted within the gate width *G*. The triples gate fraction f_t is calculated from the doubles and triples rates measured with a ^{252}Cf reference source (the parameters are defined in Section 3):

$$f_t = \frac{3f_d\nu_{s2}T}{\epsilon\nu_{s3}D} \quad (2)$$

The triples gate fraction is close to the square of the doubles gate fraction, but not exactly equal unless the counter has a single exponential die-away time and the item to be measured satisfies the assumptions of the point model.

10.5 Set the parameters for the variable-multiplication bias correction in the analysis software. This will correct multiplicity assays for the nonuniform probability of fission inside large metal plutonium items. The correction factor has the form

$$CF = 1 + a(M - 1) + b(M - 1)^2 \quad (3)$$

where *M* is the sample multiplication, and the coefficients are determined empirically or by Monte Carlo calculation. An empirical set of coefficients appropriate for metal items in several different multiplicity counters is a=0.07936 and b=0.13857 **(18)**. The correction factor approaches 1 as *M* approaches 1, so it can be left on even if the multiplicity counter is only used to assay non-metallic items, or only small metal items. Or, it can be turned off by setting a=0 and b=0 in the analysis software.

10.6 Provide physical standards for calibration, if available. Although the use of standards is not essential, the accuracy or reliability of the measurements can be increased. A complete set of standards would consist of the following:

(1) A series of ^{252}Cf sources of known isotopics and known relative strength that are referenced to a national standard, for deadtime measurements,

(2) A ^{252}Cf source or small metal Pu standard referenced to a national standard for determination of efficiency and gate fractions,

(3) A plutonium oxide standard, preferably referenced to a national standard if available, for adjustment of the triples gate fraction, and

(4) A large Pu metal standard to normalize or verify the variable-multiplication correction if Pu metal is to be measured.

(5) It is conservative, but not essential, to have additional physical standards whose plutonium mass loadings span the range of loadings expected in the items to be assayed.

If one or more representative physical standards are available, the calibration can be improved by following the steps described below.

10.6.1 Adjust the measured triples gate fraction f_t to obtain the best assay results for the standards. This corrects for uncertainties in the nuclear data parameters of ^{252}Cf and plutonium, and for differences between the actual items to be assayed and the assumptions of the point model. The adjustment to f_t may be on the order of 10 %.

10.6.2 If the *M* or α values of the physical standards are known, it may be helpful to vary ε or f_d also and obtain the best agreement with the known *M*, α, and mass values. This approach can only be helpful if the *M* or α values are well known. Otherwise, the procedure will introduce a bias into the assay of actual items that will increase as *M* or α increases.

10.6.3 As a general guideline, if there is no independent information on the *M* or α values of the standards that would provide a physical basis for adjustment, changes to the gate fractions are generally not advisable.

10.6.4 If additional calibration standards are available that are not needed to optimize the efficiency or gate fraction settings, these can be used to validate the calibration process to ensure that correct assay values are obtained on known standards.

10.6.5 When the calibration process is completed, verify the applicability of the multiplicity counting technique by measuring a series of materials to which the technique is going to be

applied. The measurements need to be verified relative to calorimetry or some other established performance comparison process.

10.7 The multiplicity calibration procedure does not need to be repeated unless there is a significant change to the physical configuration of the counter, new electronics are installed, or measurement control limits cannot be maintained. If new material categories need to be measured that may not be appropriate for multiplicity counting, some fraction of the measurements should be verified relative to calorimetry or some other established performance comparison process. For example, the ratio of counts in the inner and outer detector rings is a good indicator for neutron energy spectrum shifts that may bias the assay.

11. Measurement Control

11.1 Measurement control procedures shall be implemented to verify proper operation of the multiplicity counter. These procedures are installation specific and should be determined according to facility needs. Some of these procedures should be conducted on a daily basis, and records should be maintained to archive and monitor the measurement control results and to provide a basis for decisions about the need for re-calibration or maintenance. References **(23, 24)** describe these tests.

11.2 The quality-control tests that are commonly implemented usually include a checksum test on the shift register electronics, the accidentals/singles test, an outlier test which rejects runs that lie outside a limit, a measurement control chi-squared limit, a declared-minus-assay quality check limit, and a high voltage test limit. The tests should be selected as appropriate for the system hardware, and should include test limits that the operator can set. Runs that fail the test limits shall be rejected and identified as failed runs.

11.3 For all measurements, the count time should be split up into a minimum of 10 runs, with an individual length of 10 to 100 s. This makes it easier to diagnose electronic noise or instrument drift problems, and makes it possible to use quality control outlier tests. The outlier tests can reject runs with unusually large double or triple coincidence bursts due to cosmic rays.

11.4 Background runs should be done daily when the instrument is in use, or more frequently if there is reason to believe that the room background is changing significantly.

11.5 Normalization runs should be done daily, using the same item described in 9.2.3, to ensure that the counter is operating correctly. Because the ^{3}He detectors are very stable, the normalization constant is normally set to 1 (no correction), and rarely deviates by more than 0.5 %, unless one or more fast preamp/discriminator circuits fails. Due to the stability of these systems, if a statistically significant deviation from the expected value is obtained, the system should be taken out of service until the cause has been determined.

11.6 Occasional measurement of a known item or known representative standard is a good practice for long-term measurement control. This verifies system operation, data analysis, and large corrections like the variable-multiplication correction for metal.

12. Assay Procedure

12.1 Center the item both vertically and horizontally in the counting chamber if possible, to minimize position effects. Avoid placing items against the edges, where efficiency variations may affect assay results. This counting geometry should be maintained for all standards and assay items.

12.2 Select a count time sufficient to provide the desired measurement repeatability. This can be estimated from Fig. 3. Alternatively, select the software option that allows counting to a preset precision, if available. One percent RSD on the triple coincidence counts is commonly used, which typically requires 1000 to 1800 s of counting time. This will result in a final assay precision of about 1 % (1σ) for items with α less than 2, and about 20 % (1σ) for items with α close to 7 **(15)**.

12.3 Enter the item identification, isotopic composition, and declared Pu mass, if these are known. If data by other methods, such as passive coincidence counting, Known-*M*, or Known-α analysis is also desired these can be selected if available in the software, and if the appropriate calibration coefficients have been entered **(23, 24)**.

12.4 Carry out the item measurement. Appropriate personnel should review the data printout for data entry errors, quality control test failures, outlier test failures, and any unusual measured or calculated results.

12.5 The multiplicity counter's data acquisition and analysis software should compute the measured ^{240}Pu effective mass$_{eff}$. If the item's isotopic composition has been entered, the total Pu mass should be calculated from the equation:

$$Pu = \frac{m_{eff}}{(2.52 f_{238} + f_{240} + 1.68 f_{242})} \quad (4)$$

where f_{238}, f_{240}, and f_{242} are the mass fractions of the even plutonium isotopes present in the sample. The mass factions are usually obtained by analytical chemistry or by gamma –ray spectroscopy. The latter approach is described in Test Method C 1030.The coefficients 2.52 and 1.68 are the ratios of the spontaneous fission decay rates and second factorial moments. The available nuclear data on these coefficients has an RSD of about 2 to 3 % **(27)**.

12.6 If a previously declared mass value has been entered into the database, the assay Pu mass can be compared to the declared Pu mass, and the absolute and percent difference can be calculated.

13. Precision and Bias

13.1 Multiplicity counter assay precision is determined primarily by the statistical uncertainty in the singles, doubles, and triples count, and the reproducibility of sample placement. The dominant source of uncertainty usually comes from the triples, and is determined primarily by detector efficiency, die-away time, counting time, and the (α,n) rate of the sample.

13.2 The propagated assay uncertainty in the plutonium mass is usually estimated by the analysis software in one of two ways: from the statistical scatter between the short multiple runs that make up a single assay, or from theoretical estimation methods that have been benchmarked against measurements of the observed scatter **(15)**. See the supplier's user manual for details **(23, 24)**. In either case, the quoted error is not a Total Measurement Uncertainty (TMU) that includes all

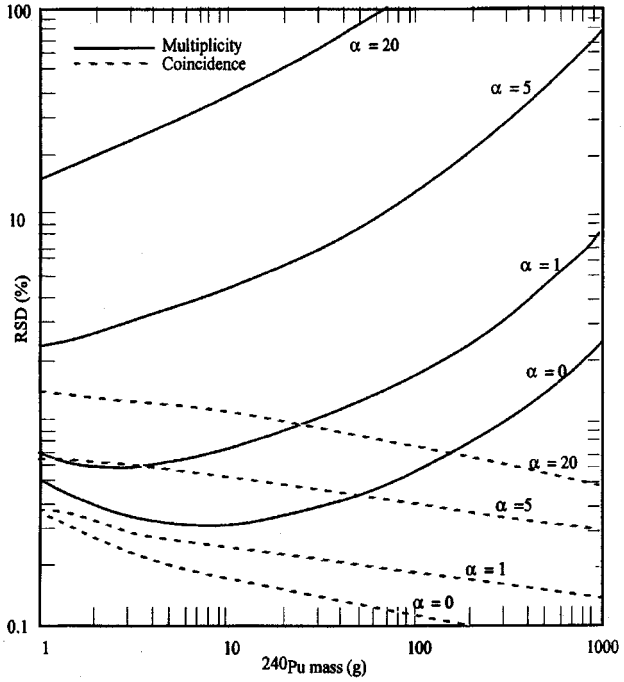

FIG. 3

Estimated precision for both multiplicity and conventional coincidence assay using a multiplicity counter with a detector efficiency of 50 %, a gate width of 64 μs, a die-away time of 50 μs, and a predelay of 3 μs. The background rate is 100 counts/s, and the counting time is 1000 s.

possible sources of error. Rather, it consists only of counting statistics and any calibration uncertainties that may be propagated.

13.3 Fig. 3 provides rough estimates of the predicted assay repeatability due to counting statistics for Pu metal (α=0), oxide (α=1), scrap (α=5), and residues (α=20) for a high-efficiency multiplicity counter **(15)**. The actual α values of such materials will vary, but the values selected here are representative. The item multiplication is estimated from typical values for plutonium oxide in cans. The curves in Fig. 3 are based on calculations that are usually within 15 to 25 % of actual observed uncertainties. Note that the repeatability due to counting statistics is always better for conventional coincidence counting than for multiplicity analysis.

13.4 Examples of single measurements of a wide range of plutonium standard cans and inventory items are given in Table 1 **(7)**. The measurements were made in a processing facility with a multiplicity counter of approximately 57 % detection efficiency and 47 μs die-away time. The measured items were in cans of 4 to 6-in. diameter, and 5 to 8-in. height. Most of the items were assayed only once, so that "precision" in this table is just the repeatability due to counting statistics. Most items were counted for 1800 s or for 3600 s, although the MSE salt was counted for 5400 s to reduce the counting statistics uncertainty due to the high α value. Except for the standards, the reference Pu total mass is based on calorimetry/isotopics, with a typical RSD of 0.6 %. The Pu-240$_{eff}$ mass fraction was

TABLE 1 Measurement Results for Multiplicity Counter Assay of Some Plutonium Items (Ref. 7)

Material Type	Pu Reference Mass (g)	Pu Assay Mass (g)	Item Multiplication M	Item alpha	Multiplicity Assay RSD	Pu240 Effective Fraction RSD	Assay Total RSD	(Assay−Reference)/ Reference (%)
Calex Std	398	394	1.102	0.9	0.2 %	[A]	0.2 %	-0.4
Oxide Std	874	877	1.084	0.7	0.8 %	[A]	0.8 %	0.3
Impure Oxide	865	855	1.074	0.9	0.6 %	2.3 %	2.3 %	-1.1
Impure Metal	2417	2463	1.483	0.0	0.3 %	3.0 %	3.0 %	1.9
Impure Metal	4074	4200	2.281	0.3	0.2 %	4.5 %	4.5 %	3.1
Pure Metal	4190	4169	2.125	0.3	0.3 %	3.6 %	3.6 %	-0.5
Filter Residue	607	626	1.057	1.6	1.5 %	1.3 %	1.9 %	3.1
MgO Crucible	130	131	1.022	2.5	0.8 %	4.0 %	4.1 %	1.0
ER Salt	493	442	1.071	3.9	0.9 %	1.9 %	2.1 %	-10.4
Sand and Slag	119	123	1.012	7.5	4.2 %	2.8 %	5.1 %	3.7
Filter Residue	339	325	1.028	10.5	9.0 %	2.6 %	9.4 %	-4.1
Impure Oxygen	314	266	1.038	30.1	44.1 %	3.0 %	44.2 %	-15.3
Inciner. Ash	161	99	1.026	30.1	47.1 %	2.0 %	47.1 %	-38.3
MSE Salt	263	188	1.021	34.2	38.9 %	3.0 %	39.0 %	-28.5

[A] Used reference isotopic values.

obtained from 1 to 2 h FRAM gamma-ray isotopic measurements, with a repeatability in the range of 1.1 to 4.6 %. The "Multiplicity Assay RSD" is the repeatability computed by the multiplicity analysis software (see 13.2). The "Pu-240 effective RSD" is the repeatability of the gamma-ray isotopic analysis of the $Pu240_{eff}$ mass fraction. The "Assay Total RSD" is the combination of these two repeatabilities in quadrature. The (Assay-Reference)/Reference uncertainty is usually within the calculated "Assay Total RSD" uncertainty. More detailed estimates of bias are given in Table 2.

13.5 Assay bias for multiplicity counting is very low for samples that meet the mathematical assumptions used in multiplicity analysis. However, in practice container and matrix factors may yield noticeable biases. Table 2 provides a broad summary of past performance for multiplicity assay of many of the nuclear materials commonly found in DOE facilities, and can be used to estimate performance for other similar applications. Table 2 also estimates the expected assay repeatability and bias (including the uncertainty from gamma-ray isotopics) relative to calorimetry or destructive analysis.

13.6 Multiplicity counting measures the even isotopes of plutonium. Biases in the determination of the plutonium isotopic composition will result in significant bias in the calculated total mass of plutonium. A fractional bias in m_{eff} propagates to the same fractional bias in the total plutonium mass.

13.7 Changes in background can affect the assay by roughly 1 % for every 1 % change in the total count rate, depending on the item's mass and self-multiplication.

13.8 The coincidence background of spallation neutrons from cosmic ray interactions can be significant for small plutonium loadings in cans with several kg of high atomic number matrix materials. For example, 100 kg of iron yields a doubles rate roughly equivalent to 20 mg ^{240}Pu, and 100 kg of lead yields a doubles rate roughly equivalent to 120 mg ^{240}Pu at 2200 m altitude. The bias is reduced to approximately one half of these values at sea level.

13.9 If the detection efficiency is not constant over the assay volume, bias effects can occur due to sample positioning or varying fill heights in the container. For a well-designed multiplicity counter these effects are usually about 1 % (1σ) for singles, 2 % (1σ) for doubles, and 3 % (1σ) for triples, and 3 % (1σ) or less for the final assay result.

13.10 The moisture content of an assay item increases α, increases self-multiplication, and alters the detection efficiency of the counter. The first two effects are calculated and automatically corrected for by the multiplicity assay, and the third can be detected by the inner/outer ring ratio if it is significant. Several wt % moisture will affect the detection efficiency of a well-designed multiplicity counter by 1 % or less.

13.11 Sample container wall effects may bias individual multiplicity assay results by about 1 % for wall thicknesses of roughly 3 to 5 mm.

13.12 Large quantities of moderator in the container can change the die-away time of the counter and bias the assay if there is no cadmium liner in the assay chamber. This effect is too counter- and matrix-specific to quantify. Monitoring the die-away time with a second gate length can provide a flag, and this option is usually available in the multiplicity hardware and software package.

13.13 Neutron poisons (at the level of several percent by weight) have no effect unless there is also enough moderator to reduce the average energy of the neutrons to the point where the poison's capture probability becomes high. At this moderator level (greater than 0.1 g/cm^3 of water or equivalent) the slower moderated neutrons tend to fall outside the coincidence counting interval. As a result, the loss of coincidence signal is no more than would be expected from the neutron detection efficiency change. This bias is seldom observed and is hard to quantify because most matrix materials do not contain large quantities of both moderator and absorber.

13.14 The presence of other spontaneous fission sources such as curium or californium will bias the assay high. For example, the spontaneous fission neutron yield from 1 mg of ^{244}Cm or 5 ng of ^{252}Cf is equivalent to the neutron output from 10 g of ^{240}Pu. If there is enough curium or californium to dominate the coincident signal, then the average observed multiplicity per fission will be higher, and the triples/doubles ratio can be used as a warning for this condition.

13.15 The response of the ^{3}He tubes, fast preamp/discriminators, and multiplicity electronics is usually stable to

TABLE 2 Summary of Past or Expected Multiplicity Counter Performance on Various Nuclear Material Categories

Nuclear Material Category	No. of Items	Ref. Technique	Pu Mass (g)	(α,n)/sf Rate α	Count Time (s)	RSD (%)	Bias (%)	Refs.
Pu Metal	13	Cal/iso	200–4000	0 to 1.3	1800	4.6	1.3	**(7)**
	14	Cal/iso	1500–5000	0	1800	2.7	-0.1	**(9)**
	5	Cal/iso	300–3700	0 to 0.3	3000	5.1	-4.7	**(26)**
Calex Std.	1	DA	398	1	1800	1.3	0.3	**(7)**
Calex Std.	1	DA	398	1	1800	1.37	0.77	**(28)**
Pu Oxide	45	Cal/iso	500–5000	1	1800	2.2	0.0	**(9)**
	5	DA	400–1800	0.7–1.1	3000	0.8	-2.7	**(26)**
Impure Pu Oxide	12	DA	20–875	0.7–4.3	1000	2–3	0.8	**(29)**
Pu Scrap	16	Cal/iso	80–1175	1–6	3600	5.7	-1.6	**(7)**
	67	Cal/iso	300–1000	1–10	1200	8	0.0	**(10)**
	24	DA	2000	1–6	1800	5.8	-1.0	**(11)**
Pu Residue	8	Cal/iso	161–339	7–34	3600	18.8	-9.2	**(7)**
	10	Cal/iso	37–300	9–32	3000	4.8	0.9	**(26)**
Mixed U/Pu Oxide	8	DA	200–800	1–2	1000	1–2	1–3	**(30)**

Note that the observed repeatability and bias estimates include the uncertainties from the neutron counting, the gamma-ray isotopic analysis of the ^{240}Pu effective fraction, and the Cal/iso reference values. For the Calex standard, Ref. **(7)** reports precision (repeatability and reproducibility) on 8 measurements, and Ref. **(28)** reports a combination of precision on about 100 measurements and repeatability on about 150 measurements. Cal/iso refers to the combination of calorimetry and gamma-ray isotopic analysis, and DA refers to destructive analysis traceable to the national measurement system.

better than 0.1 % (1σ), and contributes a negligible bias to the assay.

13.16 The average Pu mass calculated from a series of short assays may not be exactly equal to the Pu mass calculated from the average of the rates because the solution of the multiplicity analysis equations involves a non-linear cubic equation. This condition becomes more pronounced as the (α, n) rate increases, but is typically less than 0.1 %.

13.17 Care must be taken to ensure that all sources of uncertainty are included in the final reported mass value. There are several uncertainties that may not be calculated by the data analysis software, including the following:

(1) About 2 % (1σ) uncertainties in the nuclear data coefficients used to solve the multiplicity equations (These have very little effect because the calibration process compensates for them.)

(2) About 1.5 % (1σ) uncertainties in the strength of NIST-traceable ^{252}Cf sources (This will affect the assay by about 1.5 %, unless a physical standard is available to remove the uncertainty.)

(3) About 5 % (1σ) uncertainty in the variable-multiplication bias correction (For assay of large metal items, this will introduce an uncertainty of about 5 % into the assay because no large metal standards are available.).

13.18 Final assay bias for multiplicity counting is typically in the range of 1 to 5 % (1σ), as summarized in Table 2.

ANNEX

(Mandatory Information)

A1. CALCULATIONS REQUIRED TO ANALYZE DATA

A1.1 There are several electronics or mathematical approaches available for multiplicity analysis, as mentioned in the Scope. For a shift register-based system, the multiplicity counter software package should carry out the data analysis steps described in this annex to determine ^{240}Pu-effective mass m_{eff}, self-multiplication M, and (α,n) reaction rate a from the measured singles, doubles, and triples count rates **(1, 23, 24)**.

A1.2 The calculations are based on several important assumptions about the process of neutron emission and detection. To the extent that actual plutonium items meet these assumptions, the measured singles, doubles, and triples rates provide an exact solution for m_{eff}, M, and α. Otherwise, some assay biases may result. The most important assumptions are the following **(1)**:

A1.2.1 It is assumed that all induced fission neutrons are emitted simultaneously with the original spontaneous fission or (α,n) reaction (superfission concept).

A1.2.2 It is assumed that the neutron detector efficiency and the probability of fission are uniform over the sample volume. This assumption is called the point-model assumption because it is equivalent to assuming that all neutrons are emitted from one point in space.

A1.2.3 It is assumed that (α,n) neutrons and spontaneous fission neutrons have the same energy spectrum, so that the detection efficiency ε, the fission probability p, and the induced-fission multiplicity ν_i are the same for both neutron sources.

A1.2.4 It is assumed that neutron capture without fission is negligible.

A1.3 The multiplicity shift register measures the foreground multiplicity distribution in the $R + A$ gate, called *f(i)*, and the background distribution in the A gate, called *b(i)*. From these multiplicity distributions, the first three factorial moments f_k and b_k are computed. (The singles rate S times f_1 is $R + A$, and S times b_1 is A. The other factorial moments are defined in Ref. **(1)**.)

A1.4 The singles rate S, or the totals rate, is the total number of trigger events that arrive at the shift register per unit time. In terms of the computed factorial moments, the doubles rate D and the triples rate T are given by:

$$D = S(f_1 - b_1) \tag{A1.1}$$

$$T = \frac{S(f_2 - b_2 - 2b_1(f_1 - b_1))}{2} \tag{A1.2}$$

A1.5 The measured singles, doubles, and triples count rates are corrected for the background values measured during the last measurement control background run, for electronic deadtimes, and for the normalization factor determined during the last measurement control bias run, if different from 1 **(1, 23, 24)**.

A1.6 The net singles, doubles, and triples rates from an actual item are given by the following point model equations **(16)**:

$$S = F\epsilon M\nu_{s1}(1 + \alpha) \tag{A1.3}$$

$$D = \frac{F\epsilon^2 f_d M^2}{2}\left[\nu_{s2} + \left(\frac{M-1}{\nu_{i1}-1}\right)\nu_{s1}(1+\alpha)\nu_{i2}\right] \tag{A1.4}$$

$$T = \frac{F\epsilon^3 f_t M^3}{6}\left[\nu_{s3} + \left(\frac{M-1}{\nu_{i1}-1}\right)[3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3}] + 3\left(\frac{M-1}{\nu_{i1}-1}\right)^2 \nu_{s1}(1+\alpha){\nu_{i2}}^2\right] \tag{A1.5}$$

where:

F = spontaneous fission rate of the item, and the other variables are defined in Section 3.

A1.7 For measurements of large mass items in small containers, the neutron detection efficiency ε is usually assumed to be a known parameter obtained from the careful measurement of a californium reference source. Then the solution for self-multiplication M is obtained by solving the following cubic equation **(1)**:

$$a + bM + cM^2 + M^3 = 0 \tag{A1.6}$$

where the coefficients are functions of *S*, *D*, and *T*:

$$a = \frac{-6T\nu_{s2}(\nu_{i1} - 1)}{\epsilon^2 f_t S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})} \quad \text{(A1.7)}$$

$$b = \frac{2D[\nu_{s3}(\nu_{i1} - 1) - 3\nu_{s2}\nu_{i2}]}{\epsilon f_d S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})} \quad \text{(A1.8)}$$

$$c = \frac{6D\nu_{s2}\nu_{i2}}{\epsilon f_d S(\nu_{s2}\nu_{i3} - \nu_{s3}\nu_{i2})} - 1 \quad \text{(A1.9)}$$

Once *M* is determined, the item fission rate *F* is given by

$$F = \frac{\left[\frac{2D}{\epsilon f_d} - \frac{M(M-1)\,\nu_{i2}S}{\nu_{i1} - 1}\right]}{\epsilon M^2 \nu_{s2}} \quad \text{(A1.10)}$$

Once *F* is obtained, the item's ^{240}Pu effective mass m_{eff} is given by:

$$m_{eff} = \frac{F}{(473\ \text{fissions/s} - g)} \quad \text{(A1.11)}$$

Also, the item's (α,n) reaction rate α is given by:

$$\alpha = \frac{S}{(F\epsilon\nu_{s1}M)} - 1 \quad \text{(A1.12)}$$

A1.8 The ^{240}Pu-effective content of the item is multiplied by the correction factor *CF* for variable multiplication bias, if this correction was installed in the software.

$$m_{eff}(\text{corrected}) = m_{eff} \times CF \quad \text{(A1.13)}$$

The ^{240}Pu effective mass m_{eff} is that mass of plutonium that would give the same doubles response from the shift register as that obtained from all the even isotopes in the actual item **(4)**.

$$m_{eff} = 252^{238}Pu + {}^{240}Pu + 1.68\,{}^{242}Pu \quad \text{(A1.14)}$$

Then, Eq 4 in Section 12 can be used to compute the total Pu content if the isotopic composition is known.

APPENDIXES

(Nonmandatory Information)

X1. OTHER MULTIPLICITY SOLUTIONS

X1.1 Solution for Mass, Efficiency, Alpha

For measurements of low Pu mass items in large containers, such as waste drums, the neutron detection efficiency ε may vary from item to item. This is because matrix materials in the large waste containers can significantly affect the outgoing neutron energy spectrum. But in this situation, it may be a good approximation to assume that sample self-multiplication *M* equals 1. Then *M* can be considered a known parameter, and we can solve Eq A1.3-A1.5 for sample ^{240}Pu effective mass m_{eff}, α, and neutron detection efficiency ε **(17)**. First, the measured values for *S*, *D*, and *T* are used to obtain α:

$$\alpha = \frac{3ST{\nu_{s2}}^2}{2D^2\nu_{s1}\nu_{s3}} - 1 \quad \text{(X1.1)}$$

Then the sample fission rate is given by:

$$F = \frac{S^2 f_d \nu_{s2}}{2D{\nu_{s1}}^2(1+\alpha)^2} \quad \text{(X1.2)}$$

and the neutron detection efficiency is given by:

$$\epsilon = \frac{S}{F\nu_{s1}(1+\alpha)} \quad \text{(X1.3)}$$

When we use multiplicity analysis to solve for detector efficiency rather than sample multiplication, the multiplicity *RSD* increases by a factor of 3 to 4 over the entire mass range, and will be 5 to 15 % at best **(12)**.

X1.2 Solution if Alpha is Known, with *M*, Mass, Efficiency as Unknowns

For certain applications, the chemical form and isotopics may be well known but the sample container may be highly variable introducing a variability to the counting efficiency. For these samples Eq A1.3-A1.5 may be solved for *M*, ε, and m_{eff} **(17)**. The solution for self multiplication is obtained by solving the following quadratic equation:

$$0 = az^2 + bz + c \quad \text{(X1.4)}$$

where the coefficients are functions of *S*, *D*, and *T*:

$$a = \left(3 - \frac{6TSf_d^2}{4f_tD^2}\right)\nu_{s1}(1+\alpha)\nu_{i2}^2 \quad \text{(X1.5)}$$

$$b = 3\nu_{s2}\nu_{i2} + \nu_{s1}(1+\alpha)\nu_{i3} - \frac{6TSf_d^2\nu_{s2}\nu_{i2}}{2f_tD^2} \quad \text{(X1.6)}$$

$$c = \nu_{s3} - \frac{6TSf_d^2\nu_{s2}^2}{4f_tD^2\nu_{s1}(1+\alpha)} \quad \text{(X1.7)}$$

The above equations are solved for *z*, and

$$M - 1 = z(\nu_{ii} - 1) \quad \text{(X1.8)}$$

$$m_{eff} = \frac{s^2 f_d}{2DF\nu_{s1}^2(1+\alpha)^2}\left(\nu_{s2} + \nu_{i2}\nu_{s1}(1+\alpha)\left(\frac{M-1}{\nu_{i1}-1}\right)\right) \quad \text{(X1.9)}$$

$$\epsilon = \frac{S}{MFm_{240}\nu_{s1}(1+\alpha)} \quad \text{(X1.10)}$$

Potentially this method may also be of use in the analysis of the biases observed in the assay of metal forms (that is, α = 0).

X2. WHEN TO USE MULTIPLICITY COUNTING

X2.1 There are many alternatives to multiplicity counting, including calorimetry, segmented gamma-ray scanning, and passive neutron coincidence techniques. Passive coincidence options (Ref. **(31)**) include nonlinear calibration curves, the known-α approach, the known-*M* approach, and self-interrogation. Multiplicity counting may or may not be the preferred approach, depending on precision, bias, or throughput requirements.

X2.2 Factors to be considered in selecting either conventional neutron coincidence or multiplicity counting vary with material type. They include plutonium mass, (α,n) reactions, available detector efficiency, self-multiplication, neutron energy spectrum effects, spatial distribution of fissile material, other matrix effects, available counting time/required precision, and container size and shape. The assay precision for multiplicity counting is always worse than the assay precision for conventional coincidence counting, but for impure items with unknown multiplication and α, the accuracy for multiplicity counting is usually much better. Other considerations for several major material types are given below:

X2.2.1 *Pu Metal*—Pure plutonium metal has $\alpha = 0$, so conventional coincidence counting will give better assays because the precision is better. If samples are thought to be pure, but not with certainty, then multiplicity counting can be used to check the conventional assay. If conventional and multiplicity results are in statistical agreement, then the conventional result can be used; if they are in disagreement, then the multiplicity result can be used. In reality, most metal samples contain some impurities, and their surface is usually oxidized. Actual α values range from 0.1 to about 1.0, which would produce unacceptable biases in conventional coincidence counting.

X2.2.2 *Pu Oxide*—Multiplicity information is not needed if the oxide is so pure that a can be calculated, and the known-α approach can determine the mass and the multiplication from the singles and doubles rates. If samples are thought to be pure, but not with certainty, then multiplicity counting can be used to check the conventional assay. If conventional and multiplicity results are in statistical agreement, then the conventional result can be used; if they are in disagreement, then the multiplicity result can be used. Most oxide samples available in DOE facilities are impure, with actual α values between 1 and 4, and multiplicity counting will be significantly more accurate than conventional coincidence counting.

X2.2.3 *Pu Scrap*—For the purpose of this test method, scrap may be defined as plutonium with α values in the range of 1 to 6. Highly multiplying impure plutonium metal items are best assayed with multiplicity counting, but items with very low multiplication and very high (α, n) rates are best assayed with conventional coincidence counting, such as the known-*M* approach. The selection of multiplicity or coincidence counting will depend on whether the lower bias in the multiplicity assay will outweigh the loss of counting precision. The conventional coincidence result usually provides an upper mass limit because this analysis undercorrects the multiplication effects.

X2.2.4 *Pu Residues*—Residues are very heterogeneous plutonium-bearing materials with α values of 10 to 30 or more. The multiplicity technique is not well suited for residues because extremely long count times are needed to get good precision on the triples.

X2.2.5 *Pu Waste*—The additional information available from multiplicity counting can flag the presence of shielding materials, detect highly multiplying samples that should not be present, or improve assay accuracy by correcting for matrix effects such as (α, n)-induced fission or detector efficiency variations. Multiplicity assay will have poor precision relative to coincidence counting, but may be more accurate because the bias caused by (α, n) induced fissions is corrected. For screening at the TRU-waste detectability limit, multiplicity counting usually does not have sufficient precision.

X2.2.6 *Mixed Pu/U Oxides*—Mixed oxides do not meet all of the assumptions used in the multiplicity mathematics, and must be assayed with caution. If the calibration coefficients appropriate for plutonium are used to assay items with a large uranium concentration relative to their plutonium content, the assay results will tend to bias low. If the coefficients are adjusted to fit a particular mixed oxide material type with a fixed U/Pu ratio, then the multiplicity performance will be good.

X2.3 Multiplicity counting can be used successfully for inventory verification. The technique provides a higher level of verification than is possible with conventional coincidence counting because it requires less initial information about the inventory.

X2.3.1 For inventory verification, it is helpful to segregate items into categories such as calibration and measurement control standards, plutonium metal, low α plutonium (impure oxides and scrap), and high-α plutonium (residues with $\alpha > 7$). These categories can be defined by the observed assay results for sample multiplication, mass, α, or measurement precision.

X2.3.2 For low-α plutonium, counting times of 1800 s (half hour) are usually sufficient to eliminate counting statistics as a significant contribution to the overall assay precision. It may be helpful to do all assays at 1800 s, then decide on the basis of the observed αwhether additional counting time is warranted.

X2.3.3 For high α plutonium, multiplicity counting may not be a viable option because of the long count times required. Other techniques such as calorimetry, gamma-ray scanning, or the Known-*M* coincidence technique **(31)** may be preferable.

X2.3.4 The overall assay precision of multiplicity counting for total plutonium mass has a lower limit of about 3 % RSD once the error on the ^{240}Pu-effective as determined by gamma-ray isotopics is folded in. At some facilities, the use of stream average isotopics may provide better results and can eliminate the time required to do gamma-ray isotopics.

X2.3.5 Inventory verification may require the assay of storage containers with more than one sample can. Multiple discrete sources do not satisfy the mathematical assumptions used in coincidence and multiplicity counting. However, limited experience obtained to date with 2 to 5 sample cans per

drum does not indicate any observable biases in the multiplicity assay that can be attributed to this effect.

X2.3.6 Experience to date suggests that a small fraction of the inventory will have multiplicity assays that are well outside the reasonable expected limit of error because of the presence of interferences (as described in Section 6) that are not known about. These outliers will require calorimetry and/or gamma-ray isotopics to resolve.

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