



Designation: C1500 – 08

Standard Test Method for Nondestructive Assay of Plutonium by Passive Neutron Multiplicity Counting¹

This standard is issued under the fixed designation C1500; 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 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 containers including 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 1000s of g.

1.2 There are several electronics or mathematical approaches available for multiplicity analysis, including the multiplicity 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 ²⁴⁰Pu-effective by moments-based multiplicity analysis using shift register electronics (1, 2, 3) and high efficiency neutron counters specifically designed for multiplicity analysis.

1.4 This test method requires knowledge of the relative abundances of the plutonium isotopes to determine the total plutonium mass (See Test Method C1030).

1.5 This test method may also be applied to modified neutron coincidence counters (4) which were not specifically designed as multiplicity counters (that is, HLNCC, AWCC, etc), with a corresponding degradation of results.

1.6 The values stated in SI units are to be regarded as standard. No other units of measurement are included in this standard.

1.7 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate*

and determine the applicability of regulatory limitations prior to use.

2. Referenced Documents

2.1 *ASTM Standards:*³

C1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry

C1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting

C1458 Test Method for Nondestructive Assay of Plutonium, Tritium and ²⁴¹Am by Calorimetric Assay

C1490 Guide for the Selection, Training and Qualification of Nondestructive Assay (NDA) Personnel

C1592 Guide for Nondestructive Assay Measurements

C1673 Terminology of C26.10 Nondestructive Assay Methods

3. Terminology

3.1 *Definitions:*

3.1.1 Terms shall be defined in accordance with Terminology C1673 except for the following:

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

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

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

3.1.3 *factorial moment of order, n*—this is a derived quantity calculated by summing the neutron multiplicity distribution weighted by $v!/(v - n)!$ where n is the order of the moment.

3.1.4 *induced fission neutron multiplicities (v_{i1}, v_{i2}, v_{i3}), n*—the factorial moments of the induced fission neutron multiplicity distribution. Typically multiplicity analysis will utilize

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.

Current edition approved June 1, 2008. Published July 2008. Originally approved in 2002. Last previous edition approved in 2002 as C1500–02. DOI: 10.1520/C1500-08.

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

³ For referenced ASTM standards, visit the ASTM website, www.astm.org, or contact ASTM Customer Service at service@astm.org. For *Annual Book of ASTM Standards* volume information, refer to the standard's Document Summary page on the ASTM website.

the data from fast neutron-induced fission of ^{239}Pu to calculate these moments (5, 6).

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 ^3He tubes that surround the well.

4.2 The detected neutron multiplicity distribution is processed by the multiplicity shift register electronics package to obtain the number of neutrons of each multiplicity in the (R + A) and (A) gates. Gates are pictorially depicted in Fig. 1.

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 should be 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 for which suitable precision can be attained. 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, interfering neutrons, 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 the low counting statistics

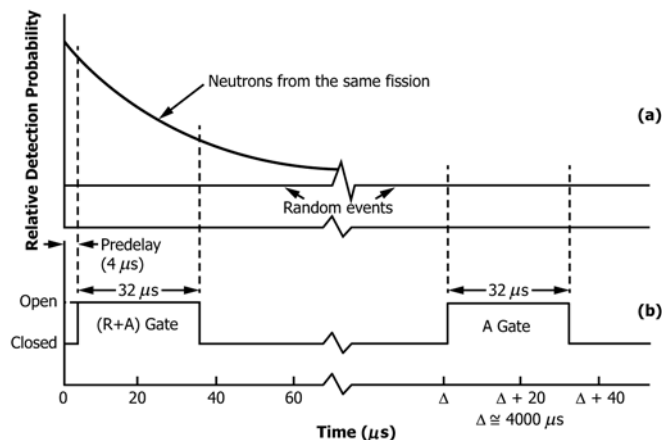


FIG. 1 (a) Simplified probability distribution showing the approximately exponential decay, as a function of time, for detecting a second neutron from a single fission event. The probability of detecting a random neutron is constant with time. (b) Typical coincidence timing parameters.

of the triple coincidences are not used. Conventional coincidence information can be obtained either by changing to coincidence analyzer mode, 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 mathematical model considered is a point in space, with assumptions that neutron detection efficiency, die-away time, and multiplication are constant across the entire item ([16](#), [17](#)). As the measurement deviates from these assumptions, the biases will increase.

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 one or more lumps that are each several hundred grams or more of plutonium metal, multiplication effects are not adequately corrected by the point model analysis ([18](#)). Variable-multiplication bias corrections must be applied.

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 (α) is low, that is, less than 10 ([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](#)).

6.3.1 High mass, high α items will produce large count rates with large accidental coincidence rates. Sometimes this prevents obtaining a meaningful result.

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 nominal levels of neutron moderation, the multiplicity analysis will automatically correct the assay for changes in multiplication. The presence of neutron poisons or other absorbers in the measurement item will introduce bias. Determination of the correction factors required for these items will have to be individually determined.

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 during measurement. This becomes important as plutonium mass decreases.

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 increase in significance 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.

6.8 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.

6.9 Unless instrument design takes high gamma-ray field into account, high gamma-ray exposure levels from the item may interfere with the neutron measurement through pile-up effects if the dose is higher than about 1 R/h at the ^3He tubes.

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 [C1207](#). Both are thermal neutron detector systems that utilize polyethylene-moderated ^3He 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. Cylindrical multiplicity well counters typically have 3 to 5 rings of ^3He tubes and absolute neutron detection efficiencies of 40 to 60 %, whereas conventional coincidence counters typically have 1 or 2 rings of ^3He 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 item positioning in the cavity is still recommended for best results.

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 ^3He 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.

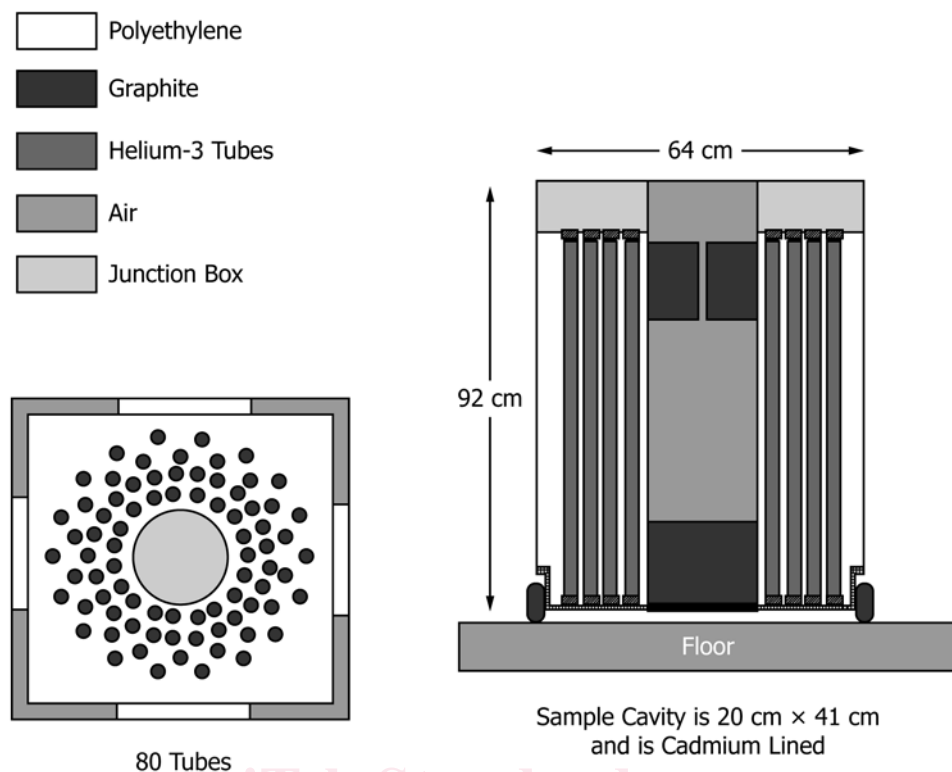


FIG. 2 Design Schematic for a Plutonium Multiplicity Counter. In this cross section of the counter, 80 ^3He tubes are arranged around the sample cavity. The space between the tubes is filled with polyethylene, and graphite above and below the sample cavity scatters and reflects neutrons. The junction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

7.2 Multiplicity Electronics:

7.2.1 An example of the physical layout of the ^3He 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 ^3He tubes require a high voltage power supply, and the electronics require a 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. If high count rates relying on the derandomizer for good results are performed, the efficacy of the derandomizer should be confirmed at the highest count rates expected.

7.2.3 A predelay circuit is usually included at the input to the multiplicity shift register to reduce the effect of small electronic transients 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 ^3He 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.

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 multiple smaller runs of equal duration.

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 ³He 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 ²⁵²Cf 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 ²⁵²Cf source is used, the ²⁵⁰Cf content should be low enough to allow decay corrections using the known half-life of ²⁵²Cf 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 ²⁵²Cf 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 ³He 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 ²⁵²Cf 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 (6, 23, 24).

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 μ s, 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 Physical standards are usually not available for a wide variety of sources and matrices. Instead, the singles, doubles, and triples equations are solved directly for multiplication M , α , and effective ²⁴⁰Pu 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 It is acceptable to use ²⁵²Cf as an experimental surrogate. 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 %.

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 ²⁵²Cf reference source (the parameters are defined in Section 3):

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

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 ²⁵²Cf reference source (the parameters are defined in Section 3):

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

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 (CF) has the form

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

where M is the item multiplication, and the coefficients a and b 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 isotopes 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_i 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_i 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 should be verified relative to calorimetric assay 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 calorimetric assay 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 ^3He 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