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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~~ 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 ~~1000's~~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 ^{240}Pu-effective by moments-based multiplicity analysis using shift register electronics **(1, 2, 3)** ~~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.~~

~~1.4This 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.~~ 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 C 1030).

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 safety and health practices and determine the applicability of regulatory limitations prior to use.*

2. Referenced Documents

2.1 *ASTM Standards:* ~~C859Terminology Relating to Nuclear Materials[3]~~
C 1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray ~~Spectroscopy[3]~~ Spectrometry
C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting
~~C1458Test Method for Nondestructive Assay of Plutonium, Tritium, and[241]Am by Calorimetric Assay[3]~~ 1458 Test Method for Nondestructive Assay of Plutonium, Tritium and 241Am by Calorimetric Assay
C 1490 Guide for the Selection, Training and Qualification of Nondestructive Assay (NDA) Personnel
C 1592 Guide for Nondestructive Assay Measurements
C 1673 Terminology of C26.10 Nondestructive Assay Methods

3. Terminology

3.1 Terms shall be defined in accordance with Terminology ~~C859~~C 1673 except for the following:

3.2 *alpha (α)*gate fractions, *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

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[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] 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* ~~, Vol 12.01.~~ volume information, refer to the standard's Document Summary page on the ASTM website.

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

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

3.3.3

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

3.4 *die-away time (τ)*

3.3 *factorial moment of order, 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.

3.9.1 *spontaneous fission neutron multiplicities (v_{s1}, v_{s2}, v_{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 v_{s1}=2.154, v_{s2}=3.789, v_{s3}=5.211 (23).

3.9.2 *induced fission neutron multiplicities (v—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.

3.4 *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 the data from fast neutron-induced fission of ^{239}Pu to calculate these moments (3). One commonly used set of moments is v_{i1}=3.163, v_{i2}=8.240, v_{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.
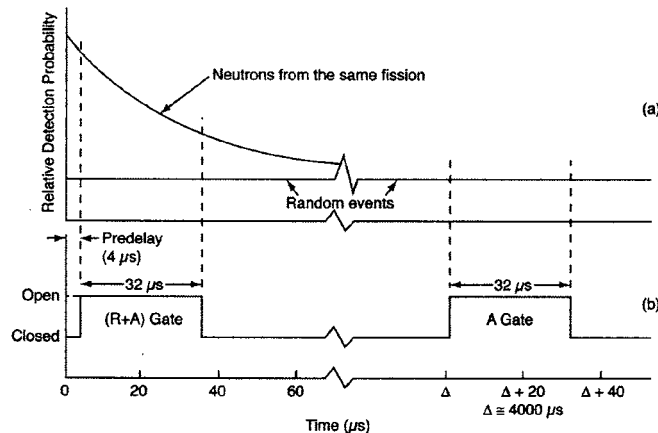

FIG. 1 (a) Simplified perobability distribution showing the approximately exponential decay, as a function of time, for detecting a second neutron from a single fission event. The probability of detecting a random neutron is constant with time. (b) Typical coincidence timing parameters.

3.11.3 *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 multiplicity shift register electronics package to obtain the number of neutrons of each multiplicity in the (R + A) and (A) gates.) 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 areshould 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, the (α,n) reaction rate,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 of the triple coincidences are not used. Conventional coincidence information can be obtained either by changing to a~coincidence counter,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 most important is the assumption that the item 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 several hundred grams of plutonium metal or more, multiplication effects are not adequately corrected by this method

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). A vVariable-multiplication bias correction is required. 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 about 7low, 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 moderatenominal levels of neutron moderation, the multiplicity analysis will automatically correct the assay for changes in multiplication. A correction for capture in The presence of neutron poisons or other absorbers is not available, so that a bias can result in measurements of such items. 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, depending on the item's mass and self-multiplication. 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 become significantincrease 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 ^{3}He 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 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. MCylindrical multiplicity well counters typically have 3 to 5 rings of ^{3}He tubes and absolute neutron detection efficiencies of 40 to 60 %, whereas conventional coincidence counters typically 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
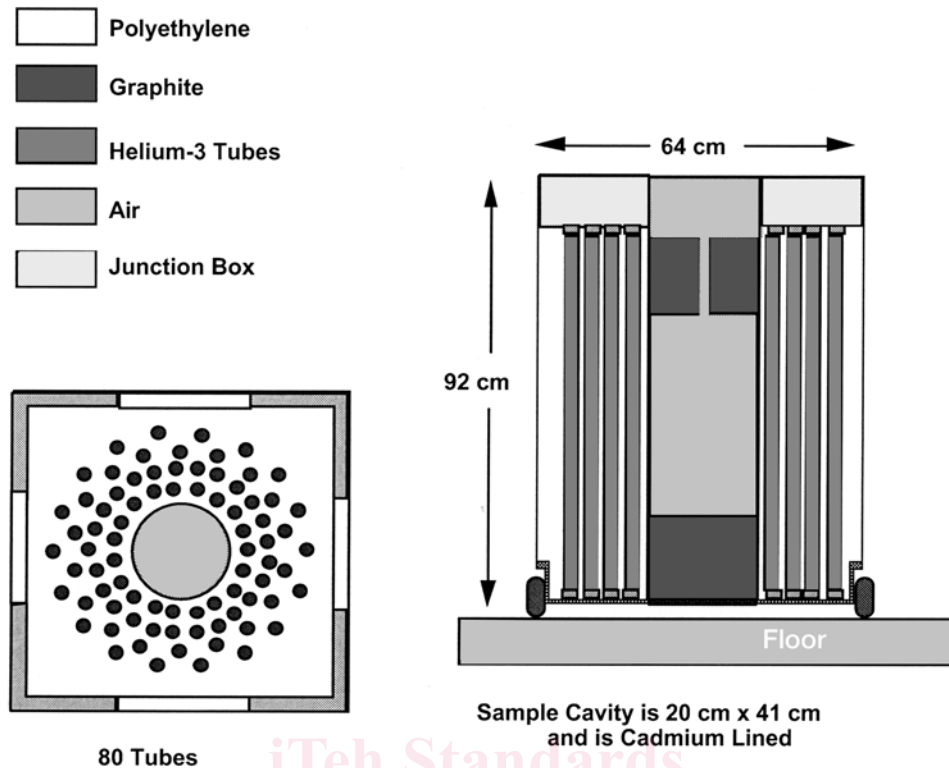
FIG. 2 Design Schematic for a Plutonium Multiplicity Counter. In this cross section of the counter, 80 ^{3}He tubes are arranged around the sample cavity. The space between the tubes is filled with polyethylene, and graphite above and below the sample cavity scatters and reflects neutrons. The junction box contains the fast preamp/discriminators.

(within several percent) to minimize the effects of item placement or item size in the cavity. Provision for reproducible ~~sample~~item positioning in the cavity is still recommended for best ~~accuracy.~~ 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 ^{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.~~

~~7.2.3A 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 .~~ *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 ^{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~~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 ^{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~~(**6, 24**~~23~~, **25**~~24~~).