Designation: C1500 – 08 (Reapproved 2017)

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

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 ^{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.

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 safety and health practices 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 ^{241}Am 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 $\nu!/(\nu - n)!$ where n is the order of the moment.

3.1.4 *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

[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 Jan. 1, 2017. Published January 2017. Originally approved in 2002. Last previous edition approved in 2008 as C1500 – 08. DOI: 10.1520/C1500-08R17.

[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 ^{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 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

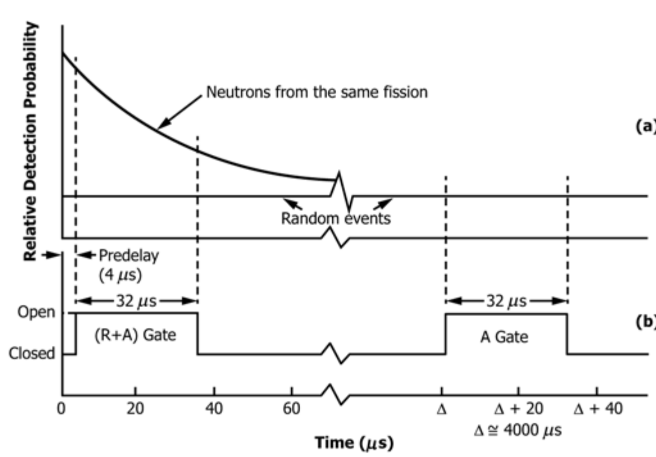

FIG. 1 (a) Simplified probability distribution showing the approximately exponential decay, as a function of time, for detecting a second neutron from a single fission event. The probability of detecting a random neutron is constant with time. (b) Typical coincidence timing parameters.

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 ^{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 C1207. 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. Cylindrical 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 (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 ^{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.