Designation: C 1493 – 01

Standard Test Method for Non-Destructive Assay of Nuclear Material in Waste by Passive and Active Neutron Counting Using a Differential Die-Away System[1]

This standard is issued under the fixed designation C 1493; 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 covers a system that performs nondestructive assay (NDA) of uranium or plutonium, or both, using the active, differential die-away technique (DDT), and passive neutron coincidence counting. Results from the active and passive measurements are combined to determine the total amount of fissile and spontaneously-fissioning material in drums of scrap or waste as large as 208 L. Corrections are made to the measurements for the effects of neutron moderation and absorption, assuming that the effects are averaged over the volume of the drum and that no significant lumps of nuclear material are present. These systems are most widely used to assay low-level and transuranic waste, but may also be used for the measurement of scrap materials. While this test method is specific to the second-generation Los Alamos National Laboratory (LANL) passive-active neutron assay system, the principle applies to other DDT systems.

1.1.1 In the active mode, the system measures fissile isotopes such as ^{235}U and ^{239}Pu. The neutrons from a pulsed, 14-MeV neutron generator are thermalized to induce fission in the assay item. Between generator pulses, the system detects prompt-fission neutrons emitted from the fissile material. The number of detected neutrons between pulses is proportional to the mass of fissile material. This method is called the differential die-away technique.

1.1.2 In the passive mode, the system detects time-coincident neutrons emitted from spontaneously fissioning isotopes. The primary isotopes measured are ^{238}Pu, ^{240}Pu, and ^{242}Pu; however, the system may be adapted for use on other spontaneously-fissioning isotopes as well. The number of coincident neutrons detected is proportional to the mass of spontaneously-fissioning material.

1.2 The active mode is used to assay fissile material in the following ranges.

1.2.1 For uranium-bearing items, the DDT can measure the ^{235}U content in the range from 0.02 to over 100 g. Normally, the assay of items bearing only uranium is performed using matrix-specific calibrations to account for the effect of the matrix on the active signal.

1.2.2 For plutonium-bearing items, the DDT method measures the ^{239}Pu content in the range between 0.01 and 20 g.

1.3 The passive mode is capable of assaying spontaneously-fissioning nuclei, over a nominal range from 0.05 to 15 g of ^{240}Pu, or equivalent. The passive mode can also be used to measure large (for example, kg) quantities of ^{238}U.

1.4 This test method requires knowledge of the relative abundances of the plutonium or uranium isotopes to determine the total plutonium or uranium mass.

1.5 This test method will give biased results when the waste form does not meet the calibration specifications and the measurement assumptions presented in this test method regarding the requirements for a homogeneous matrix, uniform source distribution, and the absence of nuclear material lumps, to the extent that they effect the measurement.

1.6 The complete active and passive assay of a 208 L drum is nominally 10 min or less.

1.7 Improvements to this test method have been reported **(1, 2, 3, 4)**.[2] Discussions of these improvements are not included in this test method.

1.8 *This standard may involve hazardous materials, operations, and equipment. 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.* Specific precautionary statements are given in Section 8.

2. Referenced Documents

2.1 *ASTM Standards:*

C 859 Terminology Relating to Nuclear Materials[3]

C 986 Guide for Developing Training Programs in the Nuclear Fuel Cycle[3]

C 1009 Guide for Establishing a Quality Assurance Program for Analytical Chemistry Laboratories within the Nuclear Industry[3]

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

[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 given in parentheses refer to a list of references at the end of the text.

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

C 1068 Guide for Qualification of Measurement Methods by a Laboratory within the Nuclear Industry[3]
C 1128 Guide for the Preparation of Working Reference Materials for Use in the Analysis of Nuclear Fuel Cycle Materials[3]
C 1156 Guide for Establishing Calibration for a Measurement Method used to Analyze Nuclear fuel Cycle Materials[3]
C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting[3]
C 1210 Guide for Establishing a Measurement System Quality Control Program for Analytical Chemistry Laboratories within the Nuclear Industry[3]
C 1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards used in the Nuclear Industry[3]

2.2 *ANSI Standard:*

ANSI N15.20 Guide to Calibrating Nondestructive Assay Systems[4]

2.3 *U.S. Government Documents:*

DOE Order 435.1 (supercedes DOE Order 5820.2A Radioactive Waste Management
DOE Order 474.1 (supercedes DOE Order 5633.3B) Control and Accountability of Nuclear Materials
DOE Order 5630.2 Control and Accountability of Nuclear Materials, Basic Principles
DOE /WIPP-069 Waste Acceptance Criteria for the Waste Isolation Pilot Plant
10CFR71 Packaging and Transport of Radioactive Materials
40CFR191 Environmental Radiation Protection Standards for Management and Disposal of Spent Nuclear Fuel, High-Level, and Transuranic Radioactive Waste
USNRC Regulatory Guide 5.11 Nondestructive Assay of Special Nuclear Materials Contained in Scrap and Waste
USNRC Regulatory Guide 5.53 Qualification, Calibration, and Error Estimation Methods for Nondestructive Assay

3. Terminology

3.1 *Definitions*—The following definitions are needed in addition to those presented in Terminology C 859.

3.1.1 *active mode*, *n*—determines total fissile mass of the assayed item through thermal neutron interrogation and subsequent detection of prompt-fission neutrons released from induced fission. A 14-MeV neutron generator is pulsed at a nominal rate of 50 Hz. The pulsed neutrons rapidly thermalize in the chamber and in the assay item. Thermal neutrons are captured by fissile material which then fissions and immediately releases more neutrons which are detected prior to the initiation of the next pulse. The prompt-neutron count rate is proportional to the mass of fissile material. This mode is called the differential die-away technique (DDT). Refer to Fig. 1.

3.1.2 *(alpha, n) reactions*, *n*—occur when energetic alpha particles collide with low atomic number nuclei such as ^{18}O, F, or Mg producing single neutrons. Neutrons produced in this manner are not correlated in time and are a source of accidentals in passive mode and background in active mode.

3.1.3 *coincidence gate length*, *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. Coincidence gate lengths are generally determined by the die-away time of the detector package. The gate length for a shielded detector package is nominally between 35 and 70 μs. The gate length for a bare detector package is nominally 250 μs.

3.1.4 *coincident neutrons*, *n*—two or more neutrons emitted simultaneously from a single event, such as from a nucleus during fission.

3.1.5 *combined passive and active*, *n*—a method which uses passive and active modes to determine the spontaneously-fissioning and fissile mass components of the waste form, respectively.

3.1.6 *depleted uranium*, *n*—uranium containing less than the naturally occurring fraction of ^{235}U isotopes (< 0.7 weight percent).

3.1.7 *die-away time*, *n*—the average lifetime of the neutron population in an NDA assay system as measured from the time of emission to detection, escape, or absorption. The average lifetime is the time required for the neutron population to decrease by a factor of 1/e. It is a function of several parameters including chamber design, detector design, assay item characteristics, and neutron energy.

3.1.8 *early gate*, *n*—the time interval during which the thermal-neutron induced prompt-fission neutrons are measured. Typically, this time interval begins 0.4 to 0.9 ms after the initiating neutron generator pulse and is 2 to 4 ms in duration. This gate is used only during the active mode. Fig. 1 indicates the approximate delay and length of the early gate in reference to a generator pulse.

3.1.9 *fissile isotopes*, *n*—isotopes that can be induced to fission by neutrons with thermal kinetic energy, about 0.025 electron volts. ^{233}U, ^{235}U, ^{239}Pu, and ^{241}Pu are the most common fissile isotopes.

3.1.10 *flux monitors*, *n*—detectors in the measurement chamber. There are two types of flux monitors:

3.1.10.1 *cavity flux monitor, n*—bare neutron detectors used to monitor the intensity of the interrogating thermal neutron flux in the chamber.

3.1.10.2 *drum flux monitors*, *n*—bare neutron detectors placed close to the drum and collimated with cadmium to measure the thermal neutron flux emitted from the drum.

3.1.11 *late gate*, *n*—the time interval during which the active neutron background is measured. Typically, this time interval begins 8 to 18 ms after the initiating neutron generator pulse. Refer to Fig. 1.

3.1.12 *lump*, *n*—that contiguous mass of nuclear material that is sufficiently large to affect the measured signal. In the active mode, self-shielding of the thermal neutron interrogating flux results in an underestimation of the fissile mass. In the passive mode, self-multiplication leads to an overestimation of the spontaneous-fissioning mass.

3.1.13 *matrix*, *n*—the material which comprises the bulk of

[4] Available from American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

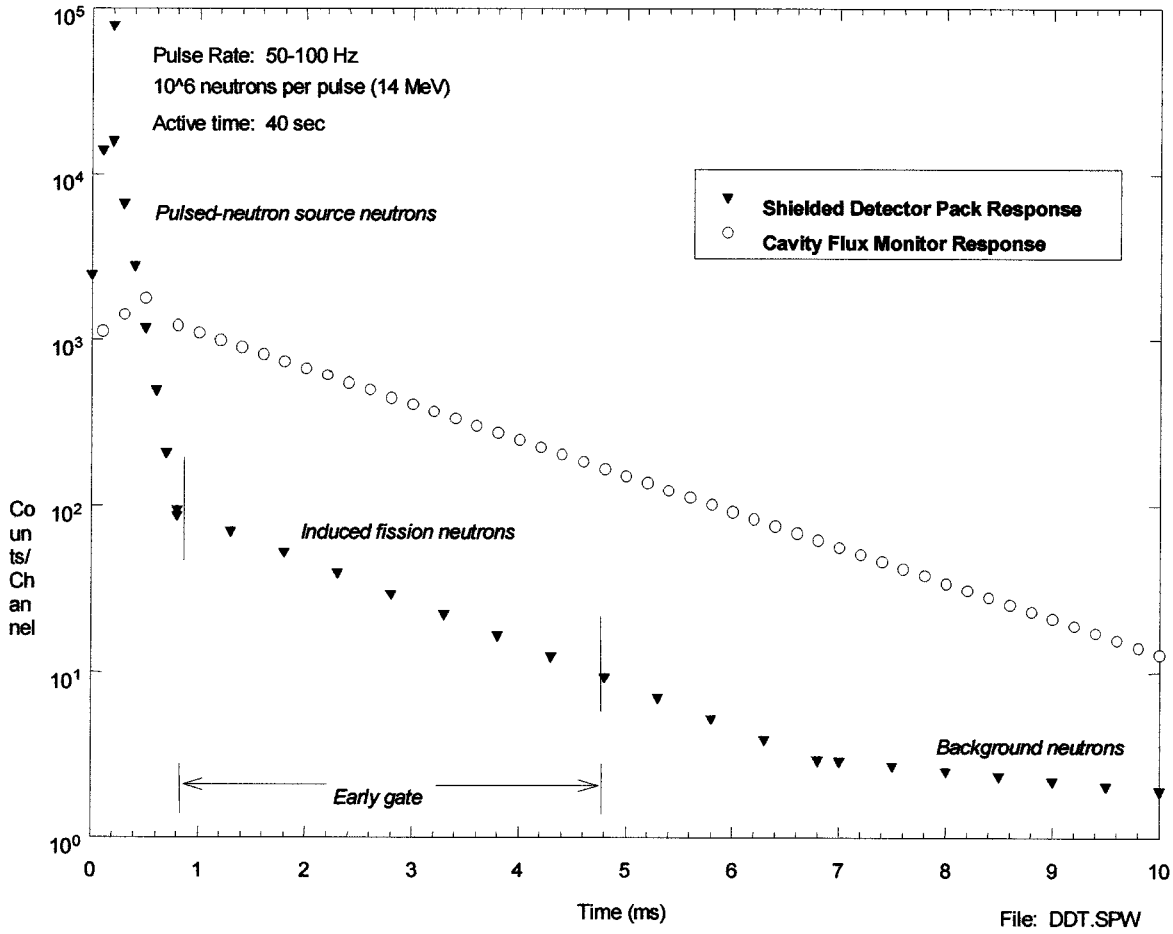

FIG. 1 Time History of an Active Assay of Plutonium Using the Differential Die-Away Technique

the item, except for the assay isotopes and the container.

3.1.14 *matrix-specific calibration, n*—uses a calibration matrix for both passive and active assays similar to the matrix to be measured. No matrix correction factors are used. This calibration is generally not appropriate for other matrices.

3.1.15 *neutron absorbers, n*—materials which have relatively large thermal-neutron capture cross-sections. Absorbers with the largest capture cross-sections are commonly known as neutron poisons. Some examples are boron, cadmium, gadolinium and lithium.

3.1.16 *neutron detector package, n*—a bundle of two or three, 2- or 3-ft long neutron proportional detectors (for example, ^{3}He tubes) surrounded by polyethylene. The output of the detectors from one package is combined into one signal and processed by a single preamplifier/amplifier/ discriminator. Neutron detector packages are of two types:

3.1.16.1 *bare detector package, n*—neutron detectors surrounded by polyethylene, but not shielded with cadmium. These packages provide a better efficiency for thermal neutrons, thus providing a better passive sensitivity when a small amount of nuclear material is present.

3.1.16.2 *shielded detector package, n*—neutron detectors surrounded by polyethylene and shielded by a thin wrapping of cadmium. These packages measure the neutrons produced by nuclear interactions and are relatively insensitive to thermalized neutrons.

3.1.17 *neutron moderators, n*—materials which slow down neutrons through elastic scattering interactions. Materials containing large amounts of low atomic weight materials, for example, hydrogen, are highly moderating.

3.1.18 *passive mode, n*—a technique used to determine the spontaneously-fissioning mass in the measured item through the detection of coincident neutrons. The coincident neutrons are prompt neutrons.

3.1.19 *^{240}Pu-effective mass, m_{eff}, n*—the mass of ^{240}Pu that would produce the same coincident neutron response in the instrument as the assay item. It is a function of the quantity of even mass isotopes of plutonium in the assay item and fundamental nuclear constants.

3.1.20 *prompt neutrons, n*—neutrons released within approximately 10^{-14} s of the fissioning event. For example, on average, ^{235}U and ^{239}Pu emit 2.41 and 2.88, respectively, prompt neutrons per neutron-induced fission event. ^{240}Pu emits on an average of 2.16 neutrons per spontaneous fission event.

3.1.21 *pulsed neutron generator, n*—a device which can supply a pulsed flux of neutrons. A widely used generator is the zetatron which produces 14-MeV neutrons via the deuterium-tritium interaction. Zetatrons generate a 10 to 20 μs pulse at a frequency of 50 Hz.

3.1.22 *spontaneously-fissioning nuclei, n*—those nuclei which do not require an external neutron source to undergo

fission. The most common isotopes are ^{238}Pu, ^{240}Pu,^{242}Pu, ^{244}Cm and ^{252}Cf.

3.1.23 *totals, n*—total number of individual neutrons detected during the count time, *t*.

3.1.23.1 *bare totals, n*—is the sum of neutrons detected from all bare detector packages.

3.1.23.2 *shielded totals, n*—is the sum of neutrons detected from all shielded detector packages.

3.1.23.3 *system totals, n*—is the sum of neutrons detected in both the bare and shielded detector packages.

3.1.24 *transuranic waste, TRU waste, n*—as defined by DOE Order 435.1 **(5)**, transuranic waste is radioactive waste containing alpha-emitting isotopes with atomic number greater than 92, half-life greater than 20 years, and with activity concentration greater than 100 nCi per gram of waste at the time of the assay.

3.1.25 *volume weighted average response, n*—an estimate of the count rate that would be obtained from a drum containing a uniform distribution of special nuclear material. It is a weighted average calculated from a series of measurements as follows: the drum is divided into typically 10 to 15 volume elements, a point source is centered in one of the volume elements and measured, the point source is moved to the next volume element and measured, and each response is weighted by the size of the corresponding element. See Appendix X1 for a more detailed explanation.

3.1.26 *wide-range calibration, n*—a calibration technique for both passive and active assays that uses calibration sources in a variety of homogeneous mock matrices whose moderation and absorption properties span the range expected to be encountered in the waste drums for which the calibration is to be used. Relationships between the measured quantities and correction factors are determined through empirical models. When the measured quantities for an "unknown" item fall within the range spanned during the calibration, the assay is within the calibration range. When an assay is not within the range spanned during the calibration, matrix-specific calibrations are performed to account for the unique properties of the item.

4. Summary of Test Method

4.1 This test method addresses a system that performs active differential die-away and passive neutron coincidence counting. Examples of the apparatus, data acquisition, and calculations contained in this test method are specific to the second-generation Los Alamos National Laboratory passive-active neutron assay system **(6)** but the principle applies to other DDT systems.

4.1.1 Typically, the active mode is performed prior to the passive mode. A 208 L drum is placed inside the chamber and rotated continuously during the measurement. The active mode is performed by interrogating the drum with neutrons from a pulsed neutron generator for 40 to 200 s. The passive mode is performed using a counting interval of 200 to 1000 s **(6, 7, 8)**. If the isotopic ratios as well as the relative responses are known for individual radionuclides, the active and passive modes can be used to give independent measurements of the total plutonium mass.

4.1.2 The system can also be operated only in the passive mode to measure the plutonium content of scrap or waste, or only in the active mode for measurement of uranium.

4.1.3 For waste containing both uranium and plutonium, determination of the mass of the fissile materials is performed outside of the standard DDT code.

4.1.4 In all modes, the relative abundances of the plutonium and uranium isotopes are required to determine the total plutonium and/or uranium mass.

4.2 The active assay is performed using the differential die-away technique **(6, 7, 8)**. The technique is described below and in Fig. 1.

4.2.1 A 14-MeV neutron generator is pulsed repeatedly, with a pulse width of 10 to 20 μs, usually at a frequency of 50 or 100 Hz.

4.2.2 After each pulse, the neutrons are quickly moderated to thermal energies in the polyethylene and/or graphite walls of the cavity and ultimately, in the waste matrix of the drum where they induce fission in fissile material.

4.2.3 The high energy neutrons from the generator that enter the Cd-shielded detector packages decrease in number exponentially (due to capture or escape). After about 600 to 900 μs, essentially all of the high energy interrogating neutrons have been cleared from the detector packages and the remaining interrogating flux of neutrons is at thermal energies.

4.2.4 Fissions induced by the interrogating neutron flux in the fissile material in the drum produce prompt high-energy neutrons, which are thermalized by the waste matrix and polyethylene in the walls of the sample chamber before being measured by the shielded detector packages during the early gate. Typically, the prompt neutrons are counted in this gate, nominally between 0.7 to 4.7 ms after each generator pulse (see Fig. 1, Region A). The difference between the fission neutron signal and the tail of the interrogation neutron signal gives rise to the name of the technique - differential die-away.

4.2.5 A background count is also made during the late gate (8 to 18 ms after each pulse after the moderated interrogating and induced fission neutrons have been cleared from the system (see Fig. 1, Region B). The late gate count is used to correct the early gate count for background neutrons, which are those neutrons, including delayed fission neutrons, that are not prompt fission neutrons.

4.2.6 The net number of prompt neutrons detected, normalized to the interrogating neutron flux as measured by the cavity flux monitor, is correlated to the quantity of fissile material in the drum.

4.2.7 The total nuclide mass is determined from the known relative abundances of the isotopes (Test Method C 1030) and the measured fissile mass.

4.3 The passive assay uses both shielded and bare detector packages to count accidentals and coincident neutrons from spontaneously-fissioning nuclei. Corrections are made to the counting data to account for background coincident neutrons. The number of coincident neutrons detected by the system is correlated to the mass of spontaneously-fissioning isotopes (for example, the even mass isotopes of plutonium) in the assay item (Test Method C 1207). The total plutonium mass is determined from the corrected coincidence count rates, the calibration curve correlating the corrected coincidence count

rates with the ^{240}Pu-effective mass, and the known or measured plutonium isotopic ratios (Test Method C 1030).

4.4 Correction factors that account for matrix effects in the observed count rates may be calculated using the ratios of counts from the cavity flux monitor and drum flux monitor (obtained during the active measurement), and from the shielded and bare detector packages (obtained during the passive measurement).

4.4.1 Generally, both ratios can be used to correct the active and passive assay results.

4.4.2 If there is no passive result, or if the passive count rates are very low (resulting in very poor counting statistics), the correction factor obtained from the ratio of the shielded and bare detector packages is not useful. For this case, the active mode results using matrix-specific calibration factors should be used.

5. Significance and Use

5.1 This test method is useful for quantifying fissile (for example, ^{233}U, ^{235}U, ^{239}Pu and ^{241}Pu) and spontaneously-fissioning nuclei (for example, ^{238}Pu, ^{240}Pu, ^{242}Pu, ^{244}Cm, ^{248} Cm, and ^{252}Cf) in waste and scrap drums. Total elemental mass of the radioactive materials can be calculated if the relative abundances of each radionuclide are known.

5.1.1 Typically, this test method is used to measure one fissile isotope (for example, ^{235}U or ^{239}Pu).

5.2 This test method can be used to segregate low level and transuranic waste at the 60 nCi/g concentration level currently required to meet the DOE Waste Isolation Pilot Plant (WIPP) waste acceptance criterion **(6, 9, 10)**.

5.3 This test method can be used for waste characterization to demonstrate compliance with the radioactivity levels specified in waste, disposal, and environmental regulations (See NRC regulatory guides, DOE Order 435.1, 10 CFR Part 71, 40 CFR Part 191, and DOE-WIPP-069).

5.3.1 In the active mode, the DDT system can measure the ^{235} U content in the range from 0.02 to over 100 g and the ^{239}Pu content, nominally between 0.01 and 20 g.

5.3.2 In the passive mode, the DDT system is capable of assaying spontaneously-fissioning nuclei, over a nominal range from 0.05 to 15 g of ^{240}Pu, or equivalent **(6, 11, 12, 13, 14)**.

5.4 This test method should be used in conjunction with a waste management plan that segregates the contents of assay items into material categories according to some or all of the following criteria: bulk density of the waste, chemical forms of the plutonium or uranium and matrix, (alpha, n) neutron intensity, hydrogen (moderator) and absorber content, thickness of fissile mass(es), and the assay item container size and composition. Each matrix may require a different set of calibration standards and may have different mass calibration limits. The effect on the quality of the assay (that is, minimizing precision and bias) can significantly depend on the degree of adherence to this waste management plan.

5.5 The bias of the measurement results is related to the fill height, the homogeneity and composition of the matrix, the quantity and distribution of the nuclear material, and the item size. The precision of the measurement results is related to the quantity of the nuclear material, the background, and the count time of the measurement.

5.5.1 For both matrix-specific and wide-range calibrations, this test method assumes the calibration material matches the items to be measured with respect to homogeneity and composition of the matrix, the neutron moderator and absorber content, and the quantity, distribution, and form of nuclear material, to the extent they affect the measurement.

5.5.2 The algorithms for this test method assume homogeneity. Heterogeneity in the distribution of nuclear material, neutron moderators, and neutron absorbers has the potential to cause biased results **(15)**.

5.5.3 This test method assumes that the distribution of the contributing radioisotopes is uniform throughout the container and that lumps of nuclear material are not present.

5.6 Reliable results from the application of this test method require training of the personnel who package the scrap and waste prior to measurement, and of personnel who perform the measurements. Training guidance is available from ANSI N15.20, Guides C 1009, C 986 and C 1068. In some cases, site-specific requirements will dictate the packaging requirements.

5.7 Both the active mode and the passive mode provide assay values for plutonium. During the calibration process, the operator should determine the applicable mass ranges for both modes of operation.

6. Interferences

6.1 Potential sources of measurement interference include self-shielding by lumps of fissile material, unexpected nuclear material contributing to the active or passive neutron signal, non-uniform nuclear material distributions within a highly moderating matrix, heterogeneity of the matrix, excessive quantities of moderators or absorbers in the matrix, multiplication, high (alpha,n) rates, high count rates, cosmic rays, high neutron backgrounds, and source characteristics (for example, oxide or metal). In general, the largest potential source of bias for active neutron measurements results from heterogeneous distributions of fissile material within a highly moderating and absorbing matrix, while the largest for the passive mode measurements is excessive neutron absorption.

6.1.1 The techniques used in this test method cannot distinguish which isotope is generating the measured response. If more than one neutron-producing nuclide is present, the relative abundances and relative responses of those radionuclides must be known.

6.1.1.1 *Active Mode*—The presence of other fissile radionuclides will increase the induced fission neutron count rate, causing an over-estimation of the ^{235}U or ^{239}Pu content, unless a correction is made. Induced fission neutrons from ^{235}U, ^{239}Pu and ^{241}Pu are indistinguishable and, therefore, the relative contributions from each of these radionuclides cannot be determined from the active assay alone. Since the calibration factor used in the calculation is isotope specific, the resulting fissile mass will be inaccurate if the relative isotopic abundances of these isotopes are unknown **(16)**.

6.1.1.2 *Passive Mode*—Other spontaneously-fissioning nuclides (for example, curium and californium) will increase the coincident neutron count rate, causing an overestimating of the plutonium content, unless their relative isotopic abundances are known. Their presence cannot be inferred from the passive

data, but discrepancies between the passive and active results may indicate their presence. Knowledge of the waste stream may also provide information on whether such interfering isotopes might be present.

6.2 Lumps of nuclear material can exhibit self-shielding or multiplication. This effect is larger for highly moderating matrices.

6.2.1 *Active Mode (Self-Shielding)*—The nuclear material on the surface of the lump shields the inside of the lump from the interrogating neutrons. Self-shielding in lumps of fissile material can lead to severe underestimates of the fissile content derived from active assays. In principle, self-shielding effects can be significant for lumps with masses containing less than 100 mg of ^{239}Pu **(17, 18)**.

6.2.2 *Passive Mode (Multiplication)*—Three factors that strongly affect the degree of multiplication are the mass of the fissile material, lump density and lump geometry. Increases in mass that are not accompanied by changes in either density or geometry will result in an increase in the coincident count rate. In general, this increase is not incorporated into the calibration function. Lumps of nuclear material are likely to cause unknown changes in multiplication and measurement bias. This effect will be negligible unless the lumps contain a few tens of grams, or more, of fissile material **(18)**.

6.3 Assay results for waste that is inhomogeneous or has a non-uniform distribution of fissile material, can have significant errors.

6.3.1 *Active Mode*—The largest errors are likely to occur in highly moderating or absorbing matrices. Generally, non-uniform distributions of fissile material can result in larger assay errors than those resulting from heterogeneous waste matrices **(6, 19)**.

6.3.2 *Passive Mode*—The largest source inhomogeneity errors are likely to occur in highly moderating matrices **(15, 17)**. Generally, there is no way to compensate for these effects.

6.4 Moderators and absorbers in the matrix can cause a bias in the measurement results, unless a correction is made. The magnitude and direction of this bias depend on the quantity of moderator present, the distribution of the fissile material, and the size of the item. The instrument produces a non-uniform response for large containers with unknown quantities of hydrogen in the matrix. In these cases, a source at the center of the container can produce either a higher or lower response than the same source located at the surface of the container.

6.4.1 *Active Mode*:

6.4.1.1 Moderation and absorption of neutrons in the waste matrix can have a large effect on the active signal, generally larger than the effects on the passive assay.

6.4.1.2 Correction factors for these effects can be obtained from calibrations using matrix-specific waste drums (see Section 9). These calibrations are usually based on homogeneous waste matrices and uniform distributions of fissile materials throughout the matrix.

6.4.2 *Passive Mode*—Neutron moderation and absorption effects can affect passive neutron count rates. The correction factors used in the technique generally account for these effects satisfactorily for uniform fissile distributions and homogeneous matrices. In general, passive counts are less affected by these effects than are active measurements **(6, 19, 20)**.

6.5 Background neutron count rates from cosmic-ray induced spallation can degrade the measurement sensitivity and the measurement precision. High-background count rates mask the instrument response.

6.5.1 *Active Mode*—Since the neutron background is measured for the active assay during the same irradiation cycles as the fissile signal is observed, sudden changes in background levels may affect the precision of the measurement, but will not result in measurement bias since the change will be accurately determined. Such rapid changes might result, for example, from movements of neutron emitting materials near the instrument. Contributions from cosmic rays and room background neutrons are generally only important at very low fissile loadings. Spontaneous fission and (alpha, n) neutrons originating in the waste drum are usually the primary contributors to the background for active assays.

6.5.2 *Passive Mode*:

6.5.2.1 Neutron background levels should be kept as low as feasible, and should not be allowed to vary significantly due to movements of neutron sources in the vicinity of the instrument. High background neutron count rates from external sources adversely affect measurement precision and detection limits.

6.5.2.2 Cosmic rays can produce coincident neutrons. Cosmic ray effects become more significant for small amounts of plutonium in the presence of large quantities of high atomic number materials such as iron or lead. Cosmic-ray induced neutrons increase in intensity as the atmospheric pressure decreases. It is possible to continuously monitor atmospheric pressure for purposes of adjusting the background count rate **(21)**.

6.6 If count rates are so high that there is a large overlap between neutrons from different coincidence events, between random neutrons, or between coincidence-event neutrons and random neutrons, precision will be poor and results may be biased for the passive mode. This typically becomes a significant problem only at count rates above 10^3 n/s. At count rates of 2×10^3 n/s or more, the shielded coincidence rate may provide a more precise and accurate result than the totals coincidence rate.

6.7 Random neutrons from (alpha, n) reactions, generally have little, if any, effect on coincidence counting.

6.7.1 If the random neutron count rate is very high compared to the coincident neutron count rate, induced multiplication effects affect the bias of the assay **(22)**.

6.7.2 Random neutrons from (alpha,n) reactions can increase the accidentals rate thereby affecting the statistical precision of the assay.

7. Apparatus

7.1 The apparatus addressed in this test method is specific to the second-generation Los Alamos National Laboratory passive-active neutron assay system **(6)**.

7.1.1 The following components are included in all second generation DDT systems. Other components, such as conveyors for drum transport and additional flux monitors, have been incorporated into some systems.

7.2 *Counting Assembly*—See Figs. 2 and 3 for a typical counting assembly configuration. The major components are

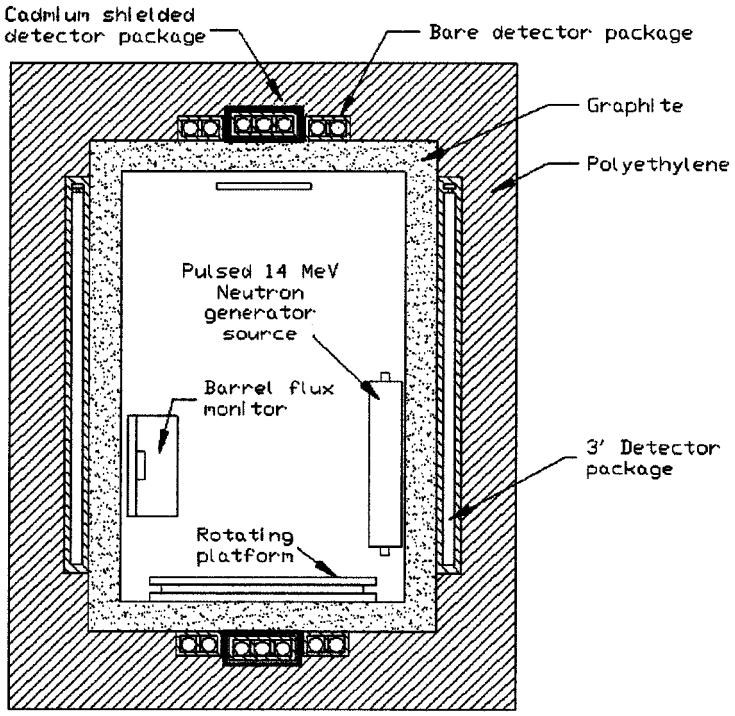

FIG. 2 Side View of DDT Counter Configuration

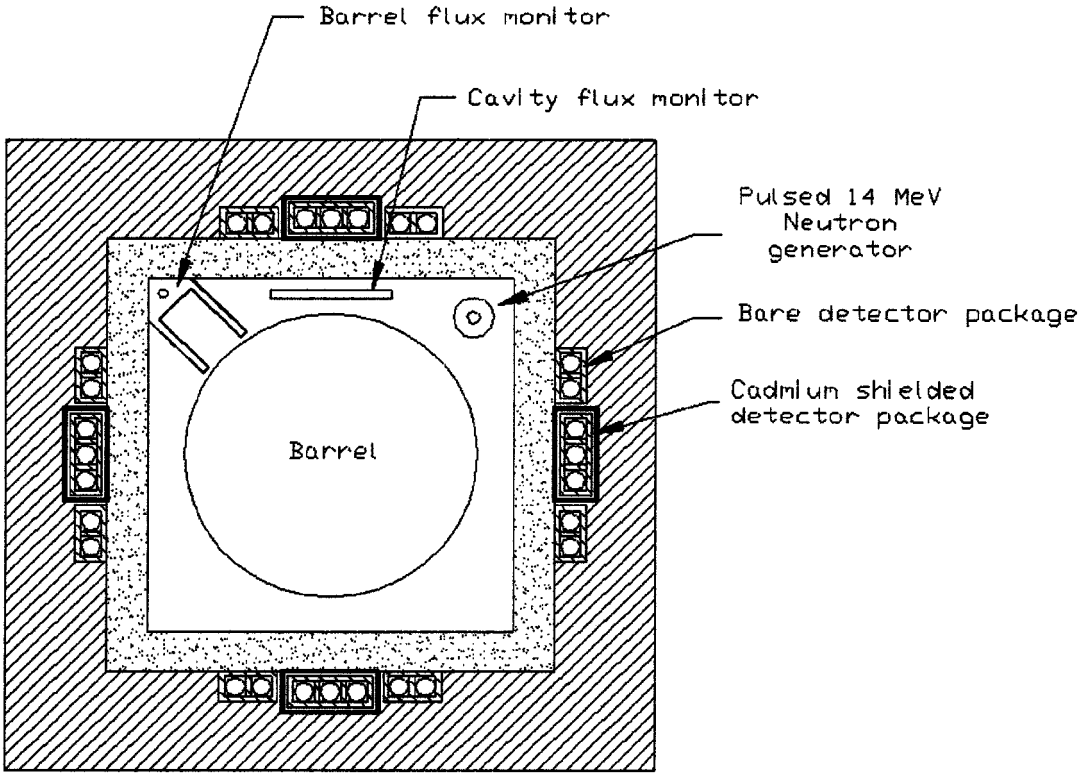

FIG. 3 Overhead View of DDT Counter Configuration

the assay chamber (polyethylene, graphite, and structural support); rotating platform; pulsed neutron source; shielded and bare neutron detector packages; cavity flux monitor; and drum flux monitor and collimator.

7.2.1 Shielded and bare neutron detector packages are positioned in the chamber walls (including the chamber door), ceiling, and floor and are used to quantify the fissioning materials in the waste (see Fig. 2).

7.2.2 The neutron detectors are embedded in polyethylene. The detection efficiency for system totals neutrons is generally between 10 and 15 % for second generation DDT systems.

7.2.3 Provision for reproducible positioning of the sample

in the chamber is important for reducing measurement bias. The same counting geometry should be maintained for the measurement of all calibration materials and assay items.

7.2.4 A 14-MeV neutron generator pulsed at 50 Hz and producing about 10^6 neutrons per pulse is generally adequate for active assays of 208 L waste drums **(23)**. The neutron generator is positioned inside the assay chamber. It provides the fast-neutron pulse which is then thermalized and used as the interrogating flux for active assays.

7.2.5 One cavity flux monitor is positioned within the assay chamber to measure the interrogating thermal neutron flux (See Fig. 3).

7.2.6 One or more drum flux monitors are positioned within the assay chamber and in close proximity to the assay item to measure the neutron flux which has elastically scattered in the assay item matrix material (see Fig. 3).

7.3 *Electronics*—Nuclear electronics convert analog pulses from the ^{3}He proportional counters to digital signals which are processed by the data acquisition system. Separate sets of preamplifiers, amplifiers, and discriminators may be provided for each detector package. The output of a discriminator is processed by scalers. The scaler outputs are manipulated by logic circuitry modules which sum the individual detector package counts **(8, 24)**.

7.3.1 Coincidence counting electronics are utilized, including a correction for accidental coincidence rates (for example, from (alpha, n) reaction neutrons and delayed fission neutrons).

7.4 Automated data acquisition and reduction are accomplished by interfacing the instrument to a computer.

7.5 *Shielding*—The assay chamber should be surrounded by layers of hydrogenous (for example, polyethylene, borated polyethylene, etc.) material to reduce neutron background caused by extraneous neutron sources and cosmic rays.

8. Hazards

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

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

8.1.2 Do not override mechanical and electrical safety systems. Precautions should be taken to minimize exposure to radiation during operation of the assay system. Plutonium, other transuranics, or fission products contained in the waste packages and the deuterium-tritium (D-T) neutron generator can produce ionizing radiation. Appropriate health physics and safety considerations should be instituted to reduce potential worker exposures.

8.1.3 Facility specific guidelines for handling and loading drums into assay system should be followed regarding criticality control.

8.1.4 The D-T neutron generator contains between 9 and 12 Ci of ^{3}H in the form of a tritiated solid. Appropriate health physics and safety considerations should be instituted to reduce potential worker exposures.

8.1.5 Shielded detector packages contain a cadmium liner. Appropriate safety considerations should be followed.

8.1.6 The system uses high-voltage electrical components. Appropriate safety considerations should be followed.

8.1.7 The neutron generator produces approximately 10^8 neutrons per second when it is operating. Although at least 2 in. of polyethylene surround the generator and shields nearby personnel from exposure to this source of radiation, appropriate safety cautions should be observed.

8.2 *Technical Hazards*:

8.2.1 Uniform neutron moderator, neutron absorber, and source distributions are assumed. Deviations may lead to biased results.

8.2.2 Locate the instrument in an area with the lowest practical neutron background. Prohibit the external movement of neutron emitters in the vicinity of the instrument while measurements are in progress.

8.2.3 Utilization of a measurement result which falls outside of the range of the calibration curve is not recommended.

8.2.4 Utilization of a result which is based on an inappropriate material category calibration is not recommended.

8.2.5 Correct operation of the neutron generator is determined by comparing the flux monitor readings with site control values. Some slow decrease in output is expected as the neutron generator tube ages, and does not affect the assay results since the shielded detector counts are normalized to the flux monitor count. Adjustment of the neutron generator may provide some compensation for this decreased output. The correct operation and adjustment of the Zetatron neutron generator is described in **(23)**.

9. Instrument Preparation and Calibration

9.1 The initial preparation of the DDT apparatus is outlined in the following sections, which discuss the initial setup, calibration, and initialization of measurement control. The details of preparation are site specific, depend on the material categories, and are generally performed by experts.

9.2 *Initial Preparation*:

9.2.1 The apparatus weight may exceed typical industrial floor load capacities. Check for adequate floor load capacity before installation.

9.2.2 Locate the apparatus to minimize radiation exposure to the operator from scrap and waste items. The DDT's shielding screens the measurement chamber from most sources of background.

9.2.3 The instrument should be located in a room where temperatures can be controlled.

9.2.4 Perform the initial setup recommended by the system manufacturer, obtaining assistance as needed.

9.2.4.1 The use of an oscilloscope to look for electronic noise during the initial setup or trouble shooting of the equipment is strongly recommended. An oscilloscope is routinely used to monitor the neutron generator source strike voltage during active counting **(23)**.

9.2.4.2 The output of the neutron generator should be checked by comparing the cavity flux monitor counts with those obtained by the manufacturer for a specified number of neutron generator pulses and specified neutron generator operating conditions (target and source voltages).

9.2.4.3 The proper operation of the instrument should be assured by performing passive and active measurements on

items containing known quantities of fissioning material. Counts and count rates of individual detectors and detector packages should be compared to those specified by the manufacturer, using appropriate scaling factors to account for differences in the strengths of the check sources.

9.3 *Preparation of Calibration Materials*—Additional sources of information include Guides C 1009, C 1068, C 1128, C 1156, C 1210, and C 1215; ANSI Guide N15.20.

9.3.1 Calibration matrices should be made from materials that will simulate the neutron moderation and absorption properties of the waste being assayed. In some cases, it may be possible to use uncontaminated material exactly the same as the waste material. Real-time Radiography (RTR) results can provide information on waste materials for mock-up drums. Often, however, it is necessary to simulate the waste matrices using a benign (non-moderating and non-absorbing) matrix material such as vermiculite, and adding various amounts of neutron moderators and absorbers, such as polyethylene beads and borax powder, respectively. The amount of moderator and absorber added to the barrels can be adjusted to approximate the properties of the real waste for which the calibration is to be used. The fabrication should document traceability for special nuclear material parameters.

9.3.2 The material in the calibration drums should be uniform throughout the barrel. Care should be exercised to avoid differential settling of components of the calibration matrices. If this is suspected to have occurred, remixing of the matrix material is recommended.

9.3.3 Because much real waste will not be composed of uniformly distributed fissile material in homogeneous waste matrices, appropriate care in the assignment of overall assay errors is required to account for differences between the calibration barrels and the actual waste composition. While it is not generally feasible to use calibration drums containing non-uniformities in matrix composition, such drums may be useful in determining the potential magnitude of errors associated with non-uniformities **(15)**.

9.3.4 Calibration sources for both the passive and active assays should span the range of loadings found in the waste barrels for which the calibration is being used. Any use of the system outside the mass range of the calibration sources should be carefully evaluated. Plutonium sources can be used for both the passive and active calibrations.

9.3.5 A source which provides both a known active and passive response is required for the calibration measurements. This may be the working standard (WS). Sources of fissile material ranging in size from nominally 10 mg to 200 g are generally required to perform a complete calibration, covering the range typically found in waste barrels. Uranium sources can be used for the active calibrations for both uranium and plutonium wastes since the relative response of these materials to thermal neutron interrogation is well known **(25)**. If uranium is used, a source of coincident neutrons, such as ^{252}Cf, is needed for the passive calibration. The relative response of the passive system to different spontaneous fission isotopes can be calculated **(26)**.

9.3.6 Both plutonium and uranium sources may exhibit significant self-shielding that must be accounted for when making an absolute calibration measurement. Self-shielding may occur in sources of very small size (mg quantities, or larger, of plutonium or enriched uranium) and multiplication can be significant in sources of 30 g, or more. These effects must be accounted for in order to properly calibrate the instrument. Typically the amount of self-shielding or multiplication is calculated using Monte Carlo techniques **(17)**.

9.4 *Calibration*:

9.4.1 The calibration of the instrument can be a lengthy and involved process (ANSI N15.20 and USNRC Guides 5.11 and 5.53). Generally, numerous measurements are made with a single source in a variety of locations in barrels containing different waste matrix materials. Additionally, several sources of different SNM masses are counted to verify the linearity, or determine the degree of non-linearity, of the system.

9.4.2 The instrument calibration may be a matrix-specific calibration, or a wide-range calibration, that is valid for a range of matrix materials and fissile loadings. The user must establish grounds to support one calibration method over another to meet individual circumstances. The wide-range calibration is normally performed by the manufacturer. If adjustments are required to electronics components, or replacements of detectors or electronics are required, the user should verify that the calibration of the system is still valid. Changes to the system electronics (for example, gate length or components) should be evaluated for their effect on the calibration.

9.4.3 Calibrations for specific matrices identified through a waste management plan described in 5.4 may lead to better results than are available in the wide-range calibration. In order to obtain the best results, packaging of the waste in each defined waste matrix category must be uniform. Each matrix category will require a set of representative calibration materials (physical standards). The effectiveness of the waste management plan and the validity of the resulting calibrations may be evaluated by monitoring the absorption and moderator indices defined below. These factors can be used to help evaluate whether the neutron emission characteristics of the calibration material match those of the assay item. Reasonable agreement between the indices for the calibration materials and assay items suggests that the calibration constants are appropriate. A facility-dependent evaluation for each matrix category is required in order to make the individual assessments.

9.4.4 *Matrix-Specific Calibration*:

9.4.4.1 If the waste being measured is made up entirely of one matrix type, such as from a waste stream where barrels are filled with a sludge of fixed composition, a calibration can be established for that specific material.

9.4.4.2 To determine the volume weighted average response, make both active and passive measurements at each of the representative positions (typically 10 to 15) in a barrel containing a matrix with the same composition as the waste stream, using appropriate calibration sources.

9.4.4.3 Combine the data for the assays obtained at the representative positions to obtain volume-weighted average correction factors for passive and active measurements. Appendix X1 summarizes the volume-weighted average approach.

9.4.4.4 Determine the passive and active responses of the

instrument as a function of mass using a series of sources that span the mass range of the waste to be assayed. This information can be used to verify the linearity of the calibration. The count rates for individual detector packages and shielded and bare combinations should be recorded . This should be performed in one fixed position in the drum.

9.4.5 *Wide-Range Calibration*:

9.4.5.1 This calibration procedure may be used to assay barrels containing materials whose neutron moderating and absorbing characteristics vary widely, as is typical of many waste streams. The objective is a calibration that will account for changes in counting efficiency and interrogation flux intensity due to varying amounts of moderators and absorbers in the waste, over a concentration range that is likely to be encountered in the waste items being assayed.

9.4.5.2 To perform this calibration, a number of drums (typically 10 to 20) containing representative homogeneous mock waste materials must be prepared. Provision must be made to place test samples of fissile material and spontaneous neutron sources at representative locations (typically 10 to 15) throughout the barrel. The matrix characteristics should span the range of moderation and absorption found in the waste to be assayed using the calibration **(6, 15)**. Any use of the system outside the range of moderation and absorption found in the calibration drums should be carefully evaluated.

9.4.5.3 For each calibration matrix, make both passive and active measurements at each representative location in the barrel. Combine the data for each barrel to obtain a volume-weighted average, which represents a uniform distribution of fissile material in the waste matrix. Appendix X1 summarizes the volume-weighted average approach.

9.4.5.4 The volume weighted system responses should then be analyzed for various parameters, such as the cavity flux monitor-to-barrel flux monitor ratio and the shielded totals to bare totals ratio, and fit to one of several mathematical functions using a fitting procedure such as the method of least squares, as described in **(6)** and **(15)**.

9.4.5.5 These fitted functions are then used to obtain matrix correction factors which are functions of measurement-derived parameters **(6, 15)**. Typically, for active measurements, one correction factor is primarily dependent on the moderating properties of the matrix, and another is primarily dependent on the matrix absorption properties. The two factors may be combined to obtain an overall active correction factor. The passive correction factor is primarily dependent on the moderation properties of the matrix.

9.4.5.6 Determine the response of the instrument as a function of mass as described in 9.4.4.4 if it has not been done previously. This check may be done with any matrix, if a suitable range of count rates can be obtained.

9.4.6 Monte Carlo neutron transport calculations may be useful in supplementing the calibration data. Such calculations may provide insight regarding the effect of matrix inhomogeneities and effects to be expected if particular materials are present in the waste matrix. Monte Carlo calculations also can be used to calculate the magnitude of the self-shielding that may be inherent in the calibration sources **(4)**.

9.4.7 The user of this test method should record the calibration procedure and data. The data should demonstrate the variation of the instrument response as a function of the nuclear mass and matrix.

9.5 *Initialize Measurement Control*—The need for adjustment of the instrument can be determined by measurement control procedures. These procedures make use of background measurements, replicate measurements of a specific item, and periodic measurement of certain items.

9.5.1 Determine the measurement control item responses and their uncertainties. These values are the ones to which future measurements will be compared (see 10.1).

9.5.2 Items used in measurement control must provide consistent measured values, within statistical expectations, each time they are measured. Perform corrections for radioactive decay when necessary.

9.5.3 Documentation of the measurement control of the instrument is required. (DOE Orders 474.1 and 5630.2)

10. Procedure

10.1 *Measurement Control*—Measurement-control measurements are made before assays of unknown samples and are interspersed between measurements of unknown samples to verify proper functioning of the instrument. If the measurement control indicates that the instrument response has changed, the operator or supervisor should determine the cause of the change and perform corrective actions. In addition, all measurements of unknown samples that have been performed since the last successful measurement control test, are suspect and may need to be repeated.

10.1.1 *Background Measurements*—Background measurements should be made in accordance with the site measurement control plan. This can be accomplished by performing, at a minimum, a passive count on an empty drum. Each detector package count or count rate can be compared with the normal background values for the package. Significant differences between background measurements may be due to such causes as electronic noise, detector or electronics failure, or to increases in neutron background count rates. Any significant discrepancy should be resolved.

10.1.2 *Bias Measurement*—Perform periodic measurements of items containing fissile material to verify the reproducibility of the instrument response in accordance with the site measurement control plan. Typical practice is to perform an active and passive count on the drum containing a measurement control item at the beginning and end of a shift for instruments used daily. For instruments that undergo intermittent use, this check is recommended before and after each use. Counts or count rates for individual detector packages, for the shielded and shielded plus bare totals, and the cavity flux monitor/drum flux monitor ratio can be compared with the values expected for that drum. Agreement with previous values within the control limits indicates long-term stability of the instrument's response. Low results may indicate that a detector or detector bank is not functioning correctly. High results may indicate electrical noise. Discrepancies should be resolved before proceeding with further measurements of unknown samples.

10.1.3 *Precision Measurement*—Perform periodic replicate measurements of items to verify the estimates of the measurement precision. The replicate measurements may be performed

either successively or using bias measurement control data performed on a daily basis. Lack of agreement might indicate background variations, electrical instabilities, or errors in the implementation of the software algorithms.

10.1.4 The data obtained above should be recorded and analyzed to determine trends in the counts or count rates for relevant parameters. These data can include: active mass (WS), passive mass (WS), system totals-to-shielded ratio (WS), cavity flux monitor/drum flux monitor ratio and cavity flux monitor response, background counts in detector sets, and responses of detectors sets (WS).

10.2 *Item Measurements*:

10.2.1 Check that the item size and container match those of the calibration standards.

10.2.2 Record the assay item (drum) information in accordance with the user's standard operating procedures.

10.2.3 The assay item should be located in the center of the assay chamber, in the same location used for the calibration. In the usual assay method, the drum is automatically rotated during measurements in order to reduce effects due to radial inhomogeneities and non-uniformities in the drum contents.

10.2.4 Measure in the active mode for the preset number of neutron generator pulses. The number of generator pulses may be set based on the desired statistical precision and may be changed to compensate for decreasing neutron generator output.

10.2.5 Measure in the passive mode for the preset counting time or until the desired statistical precision is achieved. Sufficient time should elapse between the end of the active count and the start of the passive count to ensure that there is no significant interference in the passive count from delayed neutrons resulting from the active interrogation; typically, a delay of about one minute is sufficient.

10.2.6 Calculate the fissile masses derived from the active and passive counts and calculate other quantities of interest.

11. Calculation

11.1 This section provides a brief description of the calculations developed for the second generation LANL DDT, including the calculations for calibration and for the matrix absorption/moderation correction formalism. Only major components in the analysis method are discussed as the algorithms, calibration parameters, and correction factors differ for individual systems. Reference **(6)** provides a more complete general reference. In addition, supporting documentation for individual systems should be consulted for more detailed information.

11.2 In general, the DDT algorithms, calibration parameters, and correction factors have been determined semi-empirically using simulated materials and containers that are representative of frequently-encountered waste forms.

11.3 The functional form and values of fitted parameters presented here have been put into service for some versions of the second generation of DDT systems but may not be applicable to all versions. It is the user's obligation, working in conjunction with the instrument supplier, to assure that the fitted functions, set points, and calibration schemes are suitable for the anticipated waste streams.

11.4 For waste to be measured using either wide range or matrix specific calibrations, this test method should be used in conjunction with a waste management plan as specified in Section 6. Packaging for each defined waste matrix category must be uniform. Each matrix category will require a set of representative calibration materials.

11.5 The effectiveness of the waste management plan and the validity of the calibrations are best evaluated by monitoring the absorption and moderator indices of the measured waste. These factors serve to determine whether the neutron emission characteristics of the calibration materials match those of the assayed waste. Reasonable agreement between the indices for the standards and assay items suggests that the calibration parameters are appropriate.

11.6 *Active Mode Analysis*:

11.6.1 The active neutron signal is proportional to the product of the fission cross section and the average number of neutrons per fission **(25, 26)**. These fundamental nuclear parameters are known with great confidence. This means that uranium and plutonium standards may be used interchangeably because the measured active response can be accurately adjusted between the two isotopes. Of course, this is strictly true only when self-absorption effects have been taken into account.

11.6.2 The net signal must be corrected for background and dead time effects before the fissile mass can be calculated. After corrections for these effects have been applied, fissile mass can be calculated as:

$$\textit{fissile mass} = (\textit{net signal}) \times F_A \times F_M \times F_{SA} \times C_{Active}$$

where:

- *net signal* = the net count rate (that is, corrected for background and dead time effects and normalized to the neutron generator output) in the early gate,
- F_A = the correction factor for absorption of the interrogating flux,
- F_M = the matrix correction factor for neutron moderation,
- F_{SA} = the fissile material self absorption correction factor, and
- C_A = the open geometry (empty drum, neutronically thin) active mode calibration term in units of grams of ^{239}Pu (or ^{235}U) per unit signal count.

11.6.3 To calculate total plutonium mass from fissile mass, the plutonium isotopic composition must be known.

11.6.4 For second generation DDT systems, the correction factor for absorption has generally taken the functional form **(6)**

$$F_A = C_A \times (\textit{absorption index})^{N_A}$$

where:

the Absorption Index is an empirically determined value (or average) determined from matrices that simulate a broad range of waste to be assayed, and C_A and N_A are parameters determined from a least squares fitting procedure for a number of waste matrix forms found in defense-contaminated waste. Typically, there is a threshold value below which no absorption correction is needed.

11.6.5 Absorption indices range from about 1.5 for matrices

(6) with benign materials (for example, vermiculite) to 31 for highly absorbing matrices (200 kg of water). They are calculated from the ratio of the net chamber flux monitor response to the net drum flux monitor response determined during the active mode analysis.

11.6.6 Fitting parameters of $C_A = 0.54$ and $N_A = 0.612$ have been determined as average values **(6)** over a wide variety of matrix forms. A value of approximately 2.7 has been determined as a threshold value for some second generation drum DDT systems. For DDT assays of specific waste forms, other matrix correction factors and functional forms may be more appropriate.

11.6.7 Over a broad range of matrices, the correction factor for neutron moderation has taken the form:

$$F_M = C_M \exp(N_M \times \textit{moderator index})$$

11.6.8 Again, C_M and N_M are fit from an analysis of the neutron moderation properties over a range of materials. Typical values over a wide range of waste materials are $C_M = 0.48$ and $N_M = 1.8$ **(6)**.

11.6.9 Moderator indices are measured as a function of the passive shielded totals rate, the passive totals count rate, and the absorption index. They range from 0.0 for benign matrices (vermiculate) up to 0.8 **(6)** for highly moderating materials (200 kg of water).

11.6.10 Moderator indices are not used in this form for analyses of uranium waste because uranium isotopes have only a weak passive neutron signal. No absorption correction is required for moderator indices below 0.40 **(6)**.

11.6.11 For DDT assays of specific waste forms, other neutron moderation correction factors and functional forms may be more appropriate.

11.6.12 The correction factor for self-absorption, F_{SA}, is a difficult correction to establish because it presupposes knowledge **(6)** of how the fissile material is distributed within the waste matrix. In some cases, a simple one-parameter exponential model for self-absorption has been used. The single parameter was based on an average self-absorption term determined from a number of waste drums at the Hanford facility. This correction has not been accepted by all users of second generation DDT systems.

11.6.13 Because active mode calibrations are typically a linear function of fissile mass, determination of the calibration parameter C_{Active} can often calculated from measurements of a single source. A volume-averaged source response is generally used to establish C_{Active}.

11.6.14 Since active neutron analysis uses thermal neutron interrogation of the fissile constituents in the waste, care must be taken to ensure that materials used to determine the calibration term C_{Active}, do not display self-absorption or that self-absorption corrections are calculable.

11.7 Passive neutron analysis is generally dominated by the spontaneously fissioning isotopes of plutonium: ^{238}Pu, ^{240}Pu, and ^{242}Pu. The functional form most often used to calculate the mass of the spontaneously fissioning isotopes, M_{SF}, of plutonium is

$$M_{SF} = \textit{net passive coincidence signal} \times F_P \times C_{Passive}$$

where:
the net passive coincidence signal has been corrected for background and dead time effects,
F_P = the passive mode matrix correction factor, and
$C_{Passive}$ = the passive coincidence calibration factor(s) in open geometry.

11.7.1 Again, the determination of total plutonium mass requires knowledge of the plutonium isotopic composition.

11.7.2 Over a broad range of matrix materials, passive matrix correction factors F_P, have been calculated from the Moderator Index defined above and from both the passive coincidence signal and the total neutron signal depending on the value of the Moderator index.

11.7.3 For DDT assays of specific waste forms, other matrix correction factors may be more appropriate.

11.7.4 Because most forms of plutonium have relatively weak emission rates (approximately 30 spontaneous fissions per gram per second for weapons grade plutonium) and because DDT systems have low coincidence neutron detection efficiencies (about 2 % for drum DDT systems), relatively large quantities of plutonium mass are required for the determination of the calibration parameter $C_{Passive}$. Because elaborate safeguards and security procedures are required for large masses of plutonium, ^{252}Cf standards are often used for passive mode calibration. Cross calibration correction factors that account for the different number of neutrons per fission and neutron energies for plutonium and ^{252}Cf must be used for these calibrations.

11.7.5 In contrast to active mode calibration, several plutonium calibration standards may be needed for passive mode calibration in order to correct for multiplication effects **(6)**.

12. Precision and Bias

12.1 Precision and bias are affected by many interrelated factors. These factors include neutron moderator and absorber effects; nuclear material mass; chemical form of the plutonium or uranium; bulk density and distribution of the matrix; isotopic composition; self-shielding; neutron multiplication; count rate losses; and background considerations. Evaluation of the measurement uncertainty is not purely a mathematical task, but also requires detailed knowledge of the measurement methods and the pertinent characteristics of the items being measured. Measurement of uncharacterized drums (little or no a priori knowledge of drum contents; e.g. isotopics, source and matrix distribution) may be considered to be of indeterminate accuracy because factors that tend to bias measurements may go undetected and will not be completely accounted for using the system hardware and software.

12.1.1 Precision can normally be improved if:

12.1.1.1 The count time is increased.

12.1.1.2 The background neutron rate from (alpha,n) neutron production, cosmic ray spallation events, and emissions from waste stored near the system is decreased.

12.1.1.3 This section lists precision and bias information applicable to both passive and active mode analyses, and information that is specific only to each measurement mode.

12.2 *Passive and Active Measurements*—If only plutonium is present, both the active and passive mode measurements provide assay values for plutonium. The passive result will

generally have less bias but the active result will have better precision. The active and passive measurements are not completely independent because they use correction factors that are linked.

12.2.1 *Precision*—The precision in a DDT measurement of an item can be estimated by replicate measurements.

12.2.2 *Bias*:

12.2.2.1 The bias should be estimated for each waste type.

12.2.2.2 A comparison with another assay technique can be helpful for estimating the magnitude of the bias.

12.2.2.3 Experimental studies (for example, measurements of simulated heterogeneous waste matrices) are often helpful in estimating potential bias for various waste categories.

12.2.2.4 If the detection efficiency is not constant over the assay volume, bias effects can occur due to varying fill heights, heterogeneity, or item positioning **(28)**.

12.2.2.5 Calibration materials have assigned values for mass and isotopic ratios. A bias in an assigned value causes a bias in the calculated results.

12.2.2.6 If the net passive neutron signal is small (for example, milligram quantities of plutonium) then the moderator correction may only partially account for neutron moderation effects in the measurement.

12.3 *Active Measurements*

12.3.1 *Precision*—The % Relative Standard Deviation in repeated measurements using a second generation DDT system has been shown to be less than 5 % for active mode analysis **(6)**. The mass of fissile material used for this demonstration was approximately 0.5 g for the measurement (40 s count time).

12.3.2 *Bias*:

12.3.2.1 Biases in the range of 25 % have been reported for active measurements of plutonium and uranium performed during evaluations of system performance on containers of simulated waste **(15)**.

12.3.2.2 Self-shielding can result in large biases causing an underestimation of the SNM content. Examples are cited below.

12.3.2.3 An active result obtained from a 1.0 g plutonium metal foil approximately 1.9 cm diameter and 0.2 mm thick is biased low by approximately 70 % **(4)**.

12.3.2.4 MCNP calculations predict that a 10 mg metal sphere of 93 % enriched ^{235}U will be biased low by 65 % compared to that of a dispersed source **(28)**.

12.3.2.5 Similar calculations predict that a 100 g sphere of U_3O_8 (93 % enriched) will be biased low by 11 % compared to that of a dispersed source **(28)**.

12.3.2.6 The active background is obtained during the active measurement of the waste drum by using data collected in the late gate, and is generally not a source of bias. However, for very large neutron backgrounds, on the order of 5×10^5 n/s, biases of +0.5 g have been observed **(15)**.

12.4 *Passive Measurements*:

12.4.1 *Precision*:

12.4.1.1 Counting statistics contribute a random error of less than 1 % for a 400 s count of a drum containing 30 g of low burnup plutonium metal **(15)**. The precision is generally worse for assays of smaller quantities of plutonium.

12.4.2 *Bias*:

12.4.2.1 The bias due to the cosmic-ray background is generally not significant except at plutonium loadings near the detection limit **(29)**.

12.4.2.2 Because most waste does not contain large, dense fissile sources (~100 g or more) biases due to neutron multiplication are generally negligible when compared with other uncertainties. If multiplication does occur, then the assay result will be biased high.

12.4.2.3 In drums with low moderator content, neutron absorbing materials generally have a small effect on the passive coincidence measurement. Absorbers in combination with moderators may result in significant bias, however **(15)**.

APPENDIX

(Nonmandatory Information)

X1. VOLUME WEIGHTED AVERAGE

NOTE X1.1—Other source positional calibration methods that can be shown to be equivalent to the volume-weighted method discussed in this section, may be used.

X1.1 The cost of characterizing and storing suitable calibration materials for large sets of diverse matrices can be decreased by using the volume-weighted-average concept **(6, 15)**. The concept uses small capsules of special nuclear material and containers filled with uncontaminated matrix material to estimate the response of the instrument to different matrices **(8)**.

X1.2 The containers are generally standard 208-l (55-gl) drums, 86 cm high and 57 cm in diameter. Typically, three or four tubes that span the length of the drum are placed in the drums at mid-volume radii resulting in equal volume elements in the drum. These radial distances are preferred because they are located at the mid-volume radii of a cylinder inside two nested cylindrical shells, all with equal volumes. Responses from sources at these radii approximate average responses for the equal volumes.

X1.3 When the tubes are fixed in position, the containers are filled with uncontaminated matrix material for the calibration exercise. The selection of matrix materials is based on the neutron absorption and moderation properties of the material categories used in the sorting and waste segregation procedures. The neutron moderation and absorption properties of the matrix in each container should be uniform throughout the container.

X1.4 Small capsules containing the special nuclear material of interest or appropriate surrogates are placed inside the tubes of the sample containers at various heights during the measurements. Self-shielding of the special nuclear material in each capsule should be evaluated and minimized. Identical capsules containing different quantities of special nuclear material in similar quantities can be used to evaluate self-shielding. An alternative approach **(6)** utilizes an analytical fit to the measured responses as a function of radius at a given height.

X1.5 Each matrix container will typically have measurements made at five or seven heights in each of the three tubes. If the heights are evenly spaced and the radii *r* determined above are used, each measurement is representative of the same-sized volume element. Selection of equally sized volume elements will simplify the calculations because no weighting of the responses is required.

X1.6 After the measurements are completed, the weighted average and standard deviation of the measured results are computed. Each result is weighted by the volume element it represents. The average response is representative of the response from special nuclear material distributed uniformly throughout the container.

X1.7 Another approach is to determine volume-weighted responses using Monte Carlo neutron simulations **(30)**. Modeling should be used in combination with experimental measurements.

REFERENCES

(**1**) Hensley, D.C., "Source Imaging of Drums in the APNEA System," CONF-951091, *Proceedings of the 4th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference*, Salt Lake City, Oct. 24-26, 1995.

(**2**) Hensley, D.C., "Autonomous Matrix Identification by the APNEA System," CONF-951091, *Proceedings of the 4th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference*, Salt Lake City, Oct. 24-26, 1995.

(**3**) Lucero, R.F., and Caldwell J.T., "Technical Report on the Boxes Waste Assay System," CONF-951091, *Proceedings of the 4th Nondestructive Assay and Nondestructive Examination Waste Characterization Conference*, Salt Lake City, Oct. 24-26, 1995.

(**4**) East, L.V., and Becker, G.K., "Experience Gained from Passive/Active Neutron (PAN) Assay Measurements on 12,600 TRU Waste Drums at INEL," EGG-M-94036 preprint, presented at the NDA and NDE Waste Characterization Conference, Pocatello, Idaho, February 14-16, 1994.

(**5**) U.S. Department of Energy, Radioactive Waste Management, DOE Order 435.1.

(**6**) Caldwell, J.T., Hastings, R.D., Herrera, G.C., Kunz, W.E., Shunk, E.R., "The Los Alamos Second-Generation System for Passive and Active Neutron Assays of Drum-Size Containers," Los Alamos Formal Report LA-10774-MS, September 1986.

(**7**) Kunz, W. E., et al., "A 1-mg Sensitivity Fissile Assay System," *Proc. 3rd ESARDA Symposium, Karlsruhe*, F.R. Germany, May 6-8, 1981.

(**8**) Caldwell, J.T., Bieri, J.M., and Colarusso, A.P., "The Los Alamos Second-Generation Passive-Active Neutron Assay System—FY86 Operations Record and System Evaluation," Los Alamos Technical Report LA-Q2TN-86-106, September 1986.

(**9**) Colarusso, A. P., et.al., "Mobile Nondestructive Assay System," *Proceedings of 28th Annual INMM Meeting*, Newport Beach, CA, July 12-15, 1987.

(**10**) DOE-WIPP-069, Waste Acceptance Criteria for the Waste Isolation Pilot Plant, Rev. 5, April 4, 1996.

(**11**) Schultz, F. J., et al., Oak Ridge National Laboratory; J. T. Caldwell et al., Los Alamos National Laboratory, "First-Year Evaluation of a Nondestructive Assay System for the Examination of ORNL TRU Waste," ORNL-6007, April 1984.

(**12**) Schultz, F. J., et al., "Neutron and Gamma-Ray Nondestructive Examination of Contact-Handled Transuranic Waste at the ORNL TRU Waste Drum Assay Facility," ORNL-6103, March 1985.

(**13**) Caldwell, J. T., et al., "Application of State-of-the-Art Passive and Active Neutron Assay Technology to a Decommissioning and Decontamination Project," *Proceedings 1990 Annual Meeting of the INMM*, Los Angeles, CA, July 15-18, 1990.

(**14**) Caldwell, J. T., et al., "High-Sensitivity Bulk TRU Waste Assay Systems," *Transactions ANS* Vol 46, 311-312, June 1984, New Orleans, LA.

(**15**) Nicholas, N. J., Coop, K. L., and Estep, R. J., "Capability and Limitation Study of the DDT Passive-Active Neutron Waste Instrument," Los Alamos National Laboratory Report LA-12237-MS, April, 1992.

(**16**) Perry, R. B., Brandenburg, R. W., and Beyer, N. S., "The Effect of Induced Fission on Plutonium Assay with a Neutron Coincidence Well Counter," *Transactions of the American Nuclear Society*, 15 674 (1972).

(**17**) Coop, K. L., "Neutron Die-away Methods For Criticality Safety Measurements Of Fissile Waste," *Proceedings of the International Topical Meeting on Safety Margins in Criticality Safety*, San Francisco, Nov.26-30, 1989, pp. 143-152.

(**18**) Cogbill, M. J., and Swinhoe, M. T., "Self Shielding Factors for Neutron Energies up to 0.75 keV," Harwell Laboratory Report, UKAEA, SRPD-R126, September, 1985.

(**19**) Smith, J.R., "Matrix Effects in TRU Assays Using the SWEPP Passive/Active Neutron Assay System," EG&G Idaho Informal Report, EGG-PHY-9204, August 1990.

(**20**) Caldwell, J. T., et.al., "System Evaluation Including Assay Algorithm, Matrix Corrections, and Operational Performance of the Los Alamos Passive/Active Neutron Assay Systems," Los Alamos Technical Report N2-87-222WP.

(**21**) Brunson, G. S., and Arnone, G. J., "A New System For Analyzing Neutron Multiplicities: Characterization And Some Specific Applications," Los Alamos National Laboratory Report LA-11701-MS, Nov., 1989.

(**22**) Moss, C. E., and Caldwell, J. T., "Assay of TRU Wastes Containing (alpha, n) Sources," LA UR 86-2220, June 22, 1986.

(**23**) O'Neal, M. L., "MA165C Neutron Zetatron Operating Manual," Sandia Report SAND-0152, June, 1985.

(**24**) Kuckertz, T. H., et.al., "Making Transuranics Assay Measurements Using Modern Controllers," *Proceedings 9th ESARDA Symposium on Safeguards and Nuclear Material Management*, London UK, pages 389-393, May 1987.

(**25**) Soran, P.D., and Seamon, R.E., "Graphs of the Cross Sections in the Recommended Monte Carlo Cross Section Library at the Los Alamos Scientific Laboratory," Los Alamos Scientific Laboratory Informal Report LA-8374-MS, 1980.

(**26**) Reilly, D., Ensslin, N., and Smith, H. Jr., (Ed), "Passive Nondestructive Assay of Nuclear Materials," *NUREG/CR-5550*, U.S. Government Printing Office, 1991.

(**27**) Rinard, P. M., Coop, K. L., Nicholas, N.J., Menlove, H.O., "Comparison of Shuffler and Differential Die-Away Technique Instruments for the Assay of Fissile Materials in 55-gal Waste Drums," Los Alamos National Laboratory Report LA-UR 93-2649, July 13, 1993.
(**28**) Hsue, S. T., "Non-Destructive Assay Techniques and Associated Measurement Uncertainties," JNMM, February 1992.
(**29**) Reilly, D., Ensslin, N., Smith, H. Jr., and Kreiner, S., "Passive Nondestructive Assay of Nuclear Materials," United States Nuclear Regulatory Commission, NUREG/CR-5550, March 1991.
(**30**) Haas, F. X., Marlowe, A.A., Buchl, R.A., "Modeling the Response for the Passive/Active Neutron Counting System," EG&G Rocky Flats Plant, TD-93007, October 20, 1993.