Designation: C1493 – 09

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 C1493; 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. 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. The examples given within this test method are specific to the second-generation Los Alamos National Laboratory (LANL) passive-active neutron assay system.

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, such as kilogram quantities of ^{238}U. 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-only bearing items, the DDT can measure the ^{235}U content in the range from about 0.02 to over 100 g. Small mass uranium-bearing items are typically measured using the active mode and only large mass items are measured in passive mode.

1.2.2 For plutonium-only bearing items, the DDT method measures the ^{239}Pu content in the range between about 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 ^{240}Pu equivalent.

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 but either mode can be extended to meet data quality objectives.

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

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

1.9 *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*

[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.

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:*[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

C1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards Used in the Nuclear Industry

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

2.2 *ANSI Standard:*[4]

ANSI N15.20 Guide to Calibrating Nondestructive Assay Systems

2.3 *U.S. Government Documents:*[5]

DOE Order 435.1 (supersedes DOE Order 5820.2A) Radioactive Waste Management

DOE Order 474.1 (supersedes 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

10 CFR Part 71 Packaging and Transport of Radioactive Materials

40 CFR Part 191 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:*

3.1.1 The following definitions are needed in addition to those presented in C26.10 Terminology C1673.

3.1.2 *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.

3.1.3 *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.

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

3.1.4 *early gate, n*—the time interval during which the thermal-neutron induced prompt-fission neutrons are measured.

3.1.4.1 *Discussion*—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.

3.1.5 *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.

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 LANL passive-active neutron assay system (**5**) but the principle applies to other DDT systems.

4.1.1 Typically, the active mode is performed prior to the passive mode. A208 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 (**5, 6, 7**). 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 In all modes, the relative abundances of the plutonium and uranium isotopes are required to determine the total plutonium mass, uranium mass, or both.

4.2 The active assay is performed using the differential die-away technique (**5, 6, 7**). The technique is described below and in Fig. 1.

4.2.1 A 14-MeV neutron generator is pulsed periodically, 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 walls, graphite walls, or

[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.

[4] Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, http://www.ansi.org.

[5] Available from U.S. Government Printing Office Superintendent of Documents, 732 N. Capitol St., NW, Mail Stop: SDE, Washington, DC 20401, http://www.access.gpo.gov.

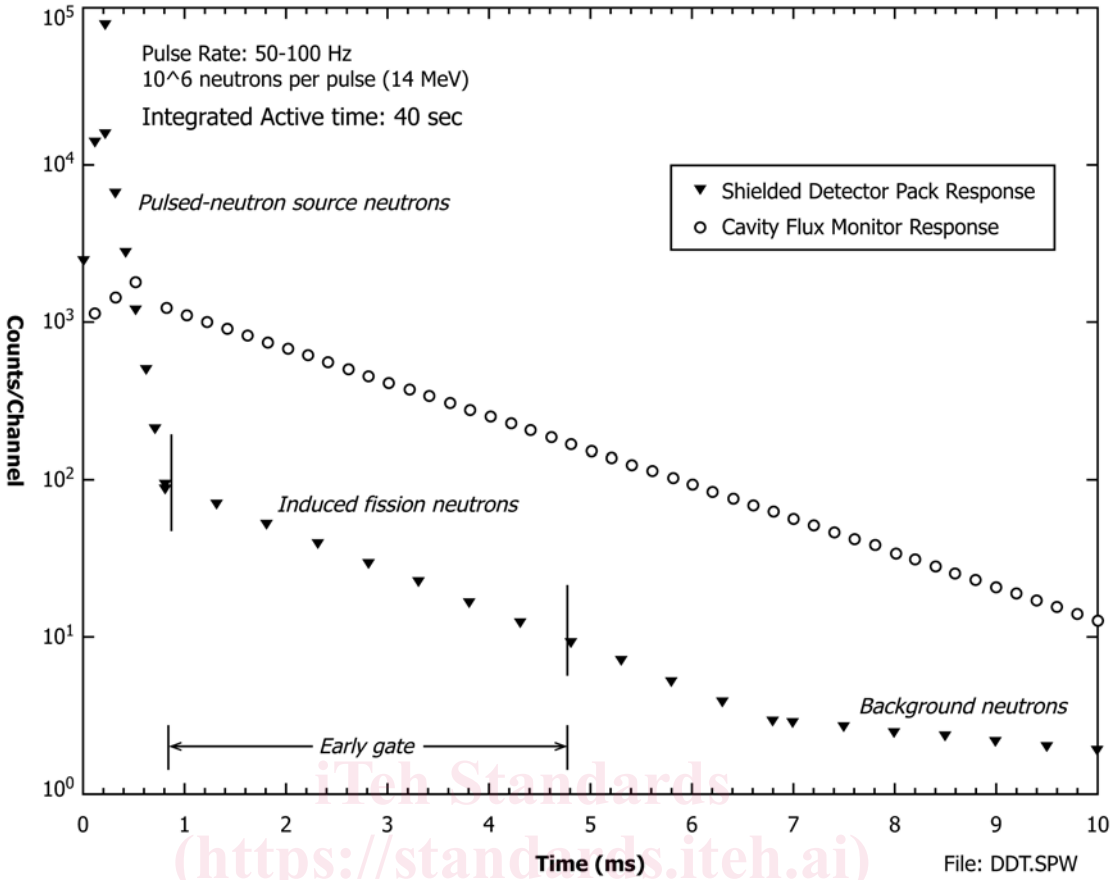

FIG. 1 Time History of an Active Assay of Plutonium Using the Differential Die-Away Technique

both, 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 the walls of the measurement 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 temporal 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 (typically 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 C1030) 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 C1207). 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 C1030).

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 100 nCi/g concentration level currently required to meet the DOE Waste Isolation Pilot Plant (WIPP) waste acceptance criterion (**5**, **8**, **9**).

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 >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 (**5**, **10**, **11**, **12**, **13**).

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, (α, 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 (**14**).

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 waste to be packaged so the conditions of Section 5.5 can be met. In some cases, site-specific requirements will dictate the packaging requirements with possible detrimental effects to the measurement results.

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:

6.1.1 self-shielding by lumps of fissile material,

6.1.2 unexpected nuclear material contributing to the active or passive neutron signal,

6.1.3 non-uniform nuclear material distributions within a moderating matrix,

6.1.4 heterogeneity of the matrix,

6.1.5 excessive quantities of moderators or absorbers in the matrix,

6.1.6 multiplication, high (α, n) rates,

6.1.7 high count rates, cosmic rays, and

6.1.8 high neutron backgrounds.

6.2 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.

6.2.1 *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 (**15**).

6.2.2 *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.

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

6.3.1 *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 (**16, 17**).

6.3.2 *Passive Mode (Multiplication)*—Four factors that strongly affect the degree of multiplication are the mass of the fissile material, (α, n), lump density and lump shape. 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 (**17**).

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

6.4.1 *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, 18**).

6.4.2 *Passive Mode*—The largest source of inhomogeneity errors are likely to occur in highly moderating matrices (**14, 16**). Generally, it is difficult to compensate for these effects.

6.5 Neutron 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.

6.5.1 *Active Mode:*

6.5.1.1 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.

6.5.1.2 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.

6.5.2 *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 (**5, 18, 19**).

6.6 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.

6.6.1 *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 (α, n) neutrons originating in the waste drum are usually the primary contributors to the background for active assays.

6.6.2 *Passive Mode:*

6.6.2.1 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 (for example, items staged on a conveyor system) adversely affect measurement precision and detection limits.

6.6.2.2 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 (**20**).

6.7 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. The shielded coincidence rate may provide a more precise and accurate result than the totals coincidence rate.

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

6.8.1 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 (**21**).

6.8.2 Random neutrons from (α, 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 LANL passive-active neutron assay system (**5**).

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 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.

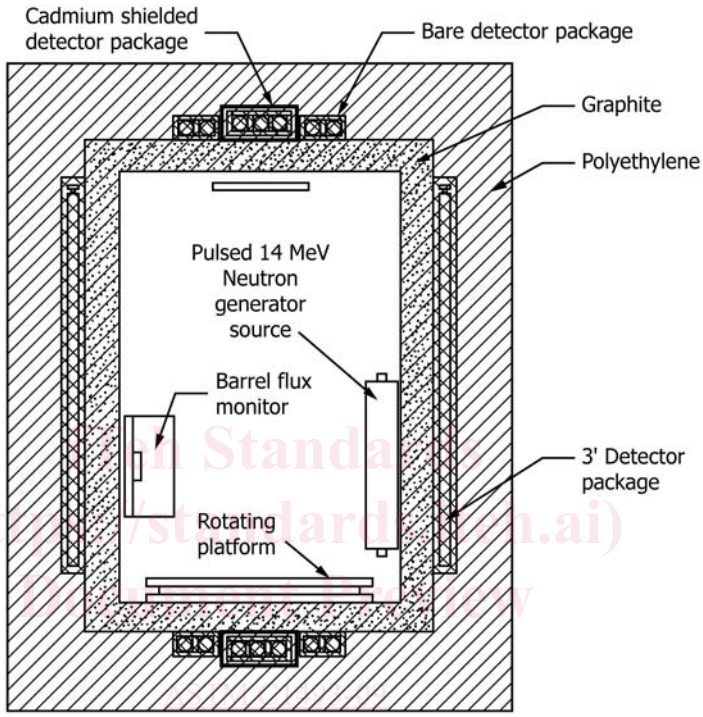

FIG. 2 Side View of DDT Counter Configuration

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 item 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 (**22**). 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