Designation: C 1316 – 08

Standard Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using ^{252}Cf Shuffler[1]

This standard is issued under the fixed designation C 1316; 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 the nondestructive assay of scrap and waste items for U, Pu, or both, using a ^{252}Cf shuffler. Shuffler measurements have been applied to a variety of matrix materials in containers of up to several 100 L. Corrections are made for the effects of matrix material. Applications of this test method include measurements for safeguards, accountability, TRU, and U waste segregation, disposal, and process control purposes (**1**, **2**, **3**).[2]

1.1.1 This test method uses passive neutron coincidence counting (**4**) to measure the ^{240}Pu-effective mass. It has been used to assay items with total Pu contents between 0.03 g and 1000 g. It could be used to measure other spontaneously fissioning isotopes such as Cm and Cf. It specifically describes the approach used with shift register electronics; however, it can be adapted to other electronics.

1.1.2 This test method uses neutron irradiation with a moveable Cf source and counting of the delayed neutrons from the induced fissions to measure the ^{235}U equivalent fissile mass. It has been used to assay items with ^{235}U contents between 0.1 g and 1000 g. It could be used to assay other fissile and fissionable isotopes.

1.2 This test method requires knowledge of the relative isotopic composition (See Test Method C 1030) of the special nuclear material to determine the mass of the different elements from the measurable quantities.

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

1.4 The techniques described in this test method have been applied to materials other than scrap and waste. These other applications are not addressed in this test method.

1.5 *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:* [3]

C 1009 Guide for Establishing a Quality Assurance Program for Analytical Chemistry Laboratories Within the Nuclear Industry

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

C 1068 Guide for Qualification of Measurement Methods by a Laboratory Within the Nuclear Industry

C 1128 Guide for Preparation of Working Reference Materials for Use in the Analysis of Nuclear Fuel Cycle Materials

C 1133 Test Method for Nondestructive Assay of Special Nuclear Material in Low-Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning

C 1156 Guide for Establishing Calibration for a Measurement Method Used to Analyze Nuclear Fuel Cycle Materials

C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting

C 1210 Guide for Establishing a Measurement System Quality Control Program for Analytical Chemistry Laboratories Within the Nuclear Industry

C 1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards Used in the Nuclear Industry

C 1490 Guide for the Selection, Training and Qualification of Nondestructive Assay (NDA) Personnel

C 1592 Guide for Nondestructive Assay Measurements

C 1673 Terminology of C26.10 Nondestructive Assay Methods

[1] This test method is under the jurisdiction of ASTM Committee C26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.10 on Non Destructive Assay.

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[2] The boldface numbers in parentheses refer to a list of references at the end of this test method.

[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.

2.2 *ANSI Documents:*

ANSI 15.20 Guide to Calibrating Nondestructive Assay Systems[4]

ANSI N15.36 Nondestructive Assay Measurement Control and Assurance[4]

3. Terminology

3.1 *Definitions*—Terms shall be defined in accordance with Terminology C 1673.

3.2 *Definitions of Terms Specific to This Standard:*

3.2.1 *active mode*, *n*—determines total fissile mass in the assayed item through neutron interrogation and counting of the delayed neutrons from induced fissions.

4. Summary of Test Method

4.1 This test method consists of two distinct modes of operation: passive and active. The instrument that performs the active mode measurement is referred to as a shuffler due to the cyclic motion of the ^{252}Cf source. This test method usually relies on passive neutron coincidence counting to determine the Pu content of the item, and active neutron irradiation followed by delayed neutron counting to determine the U content.

4.1.1 *Passive Neutron Coincidence Counting Mode*—The even mass isotopes of Pu fission spontaneously. On average approximately 2.2 prompt neutrons are emitted per fission. The number of coincident fission neutrons detected by the instrument is correlated to the quantity of even mass isotopes of Pu. The total Pu mass is determined from the known isotopic ratios and the measured quantity of even mass isotopes. This test method refers specifically to the shift register coincidence counting electronics (see **(4)** and Test Method C 1207).

4.1.2 *Active Neutron (Shuffler) Mode*—Fissions in ^{235}U, ^{239}Pu and other fissile nuclides can be induced by bombarding them with neutrons. Approximately 1 % of the neutrons emitted per fission are delayed in time, being emitted from the fission products over the time range from μs to several minutes after the fission event. Roberts et. al **(5)** were the first to observe delayed neutron emission. We now know that over 270 delayed neutron precursors contribute to the yield although the time behavior can be adequately described for most purposes using a few (six to eight) effective groups each with a characteristic time constant. The idea of detecting delayed neutrons for the analysis of ^{235}U has been attributed to Echo and Turk **(6)**. The active shuffler mode consists of several irradiate-count cycles, or shuffles, of the ^{252}Cf neutron source between the positions illustrated in Fig. 1. ^{252}Cf emits a fission neutron spectrum. During each shuffle, the ^{252}Cf source is moved close to the item for a short irradiation, then moved to a shielded position while the delayed neutrons are counted. The number of delayed neutrons detected is correlated with the quantity of fissile and fissionable material. The total U mass is determined from the known relative isotopic compostion and the measured quantity of ^{235}U equivalent **(7)**.

4.2 Either corrections are made for the effects of neutron absorbers and moderators in the matrix, or a matrix-specific calibration is used. The effect that needs correction is the increase or decrease in the specific neutron signal caused by the matrix.

4.3 Corrections are made for deadtime, neutron background, and the Cf source decay.

4.4 The active mode also induces fissions in Pu if it is present in the assay item. The passive measurement of Pu can be used to correct the active measurement of ^{235}U effective for the presence of Pu.

4.5 Calibrations are generally based on measurements of well documented reference materials **(8)** and may be extended by calculation **(9-11)**. The method includes measurement control tests to verify reliable and stable performance of the instrument.

5. Significance and Use

5.1 This test method is used to determine the U and Pu content of scrap and waste in containers. Active measurement times have typically been 100 to 1000 s. Passive measurement times have typically been 400 s to several hours. The following limits may be further restricted depending upon specific matrix, calibration material, criticality safety, or counting equipment considerations.

5.1.1 The passive measurement has been applied to benign matrices in 208 L drums with Pu content ranging from 30 mg to 1 kg.

5.1.2 The active measurement has been applied to waste drums with ^{235}U content ranging from about 100 mg to 1 kg.

5.2 This test method can be used to demonstrate compliance with the radioactivity levels specified in safeguards, waste, disposal, and environmental regulations (for example, see NRC regulatory guides 5.11, 5.53, DOE Order 5820.2a, and 10CFR61 sections 61.55 and sections 61.56, 40CFR191, and DOE/WIPP-069).

5.3 This test method could be used to detect diversion attempts that use shielding to encapsulate nuclear material.

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

5.4.1 For both the matrix-specific and the matrix-correction approaches, the method assumes the calibration materials match the items to be measured with respect to the homogeneity and composition of the matrix, the neutron moderator and absorber content, and the quantity of nuclear material, to the extent they affect the measurement.

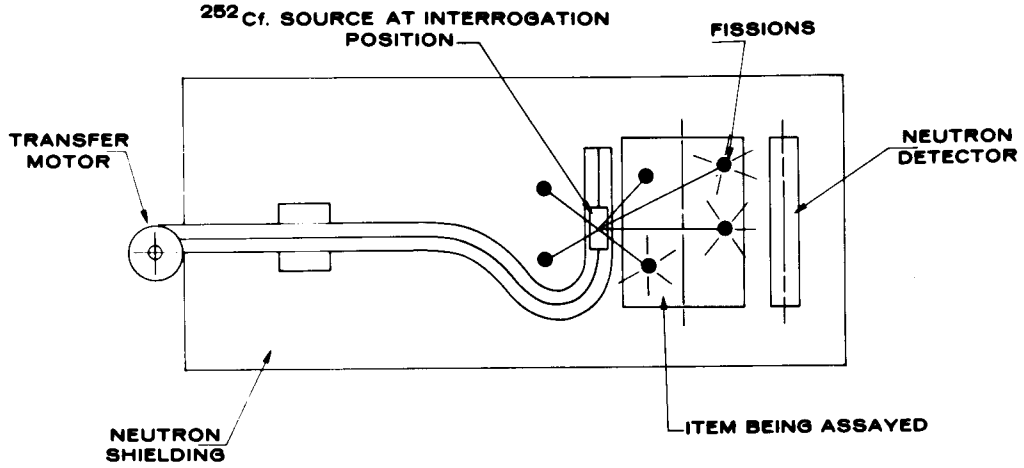
5.4.2 It is recommended that measurements be made on small containers of scrap and waste before they are combined in large containers. Special arrangement may be required to assay small containers to best effect in a large cavity general purpose shuffer.

5.4.3 It is recommended that measurements be made on containers with homogeneous contents. In general, heterogeneity in the distribution of nuclear material, neutron moderators, and neutron absorbers has the potential to cause biased results.

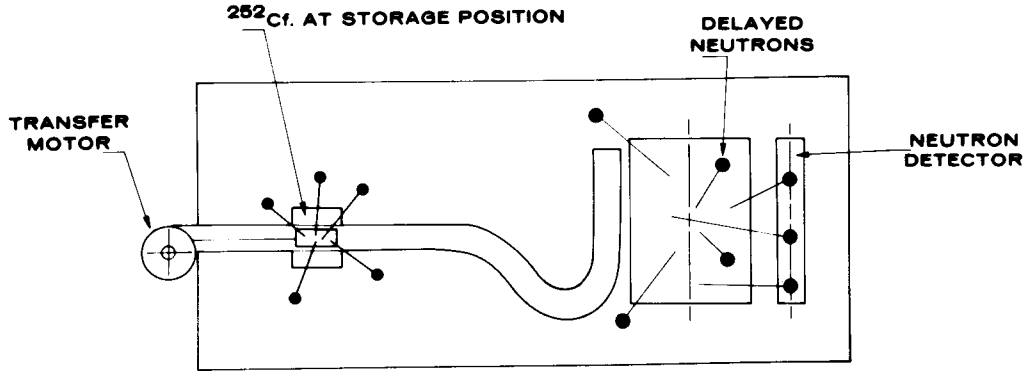
[4] Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, http://www.ansi.org.

^{252}Cf. SHUFFLER MEASUREMENT PRINCIPLE

A ^{252}Cf. NEUTRON SOURCE IS USED TO INDUCE FISSIONS IN THE SAMPLE.



DELAYED NEUTRONS ARE COUNTED WITH THE SOURCE STORED



NOTE 1—The shuffler measurement consists of several cycles. Each cycle includes the movement of the ^{252}Cf source from the storage (or home) position to the irradiation position close to the item, irradiation of the item for a period of about 10 s, return of the source to the shield followed by a counting period of about 10 s. In obvious notation this cycle structure may be succinctly described by the four time periods involved (t_{in}, t_{irr}, t_{out}, t_{cnt}). Typically the one-way transit times are less than 1 s.

FIG. 1 Cf Shuffler Measurement Principle

5.5 This test method requires that the relative isotopic compositions of the contributing elements are known.

5.6 This test method assumes that the distribution of the contributing isotopes is uniform throughout the container when the matrix affects neutron transport.

5.7 This test method assumes that lump affects are unimportant—that is to say that large quantities of special nuclear material are not concentrated in a small portion of the container.

5.8 For best results from the application of this test method, appropriate packaging of the items is required. Suitable training of the personnel who package the scrap and waste prior to measurement should be provided (for example, see ANSI 15.20, Guide C 1009, Guide C 1490, and Guide C 1068 for training guidance). Sometimes site specific conditions and requirements may have greater bearing.

6. Interferences

6.1 Potential sources of measurement interference include unexpected nuclear material contributing to the active or passive neutron signal, self-shielding by lumps of fissile material, neutron self-multiplication, excessive quantities of absorbers or moderators in the matrix, heterogeneity of the matrix, and the non-uniformity of the nuclear material spatial distribution especially within a moderating matrix. In general, the greatest potential source of bias for active neutron measurement is heterogeneity of the nuclear material within a highly moderating matrix, while the greatest for passive neutron measurement is neutron moderation and absorption (**12**).

6.2 The techniques described in this test method cannot distinguish which isotope is generating the measured response. If more than one nuclide that produces a response is present, the relative abundances and relative specific responses of those nuclides must be known.

6.2.1 *Active Mode*—The unidentified presence of other fissionable nuclides will increase the delayed neutron count rate, causing an overestimation of the ^{235}U content. For example, a calibration based on highly enriched U will cause biased results if the unknowns actually contain low-enriched U due to the potential difference in the fractional contribution arising from the fast fission in ^{238}U (**13, 14**).

6.2.2 *Passive Mode*—The unidentified presence of other spontaneous fission nuclides, such as Cm and Cf, will increase the coincident neutron rates, causing an overestimation of the Pu content. The active mode measurement of Pu is generally not sensitive to this source of bias (although counting precision may be affected) because the masses of concern are so small and present a comparatively tiny induced fission signal.

6.3 Lumps of nuclear material can exhibit self-shielding or multiplication. This effect is often larger for moderating (hydrogenous) 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 (**15, 16**).

6.3.2 *Passive Mode (Multiplication)*—Neutrons originating in the lump induce fissions in the same lump which boosts the specific coincident rate.

6.4 Moderators in the matrix can cause a bias in the measurement results, unless a correction is made or an appropriate matrix specific calibration is used. 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 (**2, 17**).

6.4.1 Although moderation is the greatest potential source of bias for passive measurements, the passive method is generally less susceptible to the presence of moderator than the active method.

6.4.2 The presence of absorbers in the matrix can cause bias if there is sufficient moderator present. The moderator slows fast neutrons which can then be captured more effectively by the absorbers.

6.4.3 The instrument produces a nonuniform response across the container, the severity varying with the concentration of hydrogen in the matrix. 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 depending on the item and instrument design.

6.5 Background neutron count rates from cosmic ray-induced spallation can degrade the measurement sensitivity (detection limit) and the measurement precision for small masses (**18, 19**).

6.6 High-background count rates mask the instrument response to small quantities of special nuclear material for both the active and passive modes (**20-22**).

6.7 High gamma dose rates eminating from the item (>10 mSv h^{-1} of penetrating radiation) may cause pile-up and break-down in the ^{3}He-filled proportional neutron detectors (**23**). Care should be taken to ensure the item is within the acceptable range of the instrument.

6.8 Certain other elements may produce delayed neutrons following (fast) neutron irradiation (**24**).

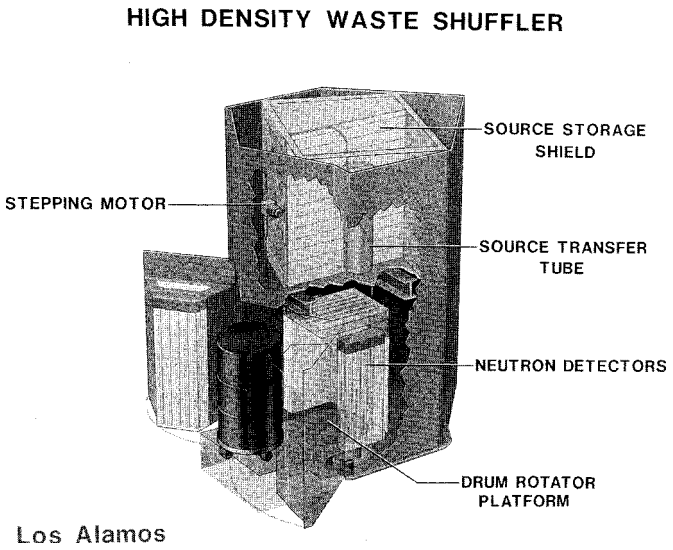
7. Apparatus

7.1 The apparatus used in this test method can be obtained commercially. Specific applications may require customized designs to cope with (for example) container sizes, container weights, activity levels, integration into the facility (**23, 25-28**). The following description is one possible design. Fig. 2 is a cutaway illustration of a shuffler to measure 208 L drums. In this design, the ^{252}Cf source storage shield is positioned on top of the measurement chamber. This design weighs approximately 8000 kg, and is 3 m high and 2 m in diameter.

7.2 *Counting Assembly*—see Fig. 3.

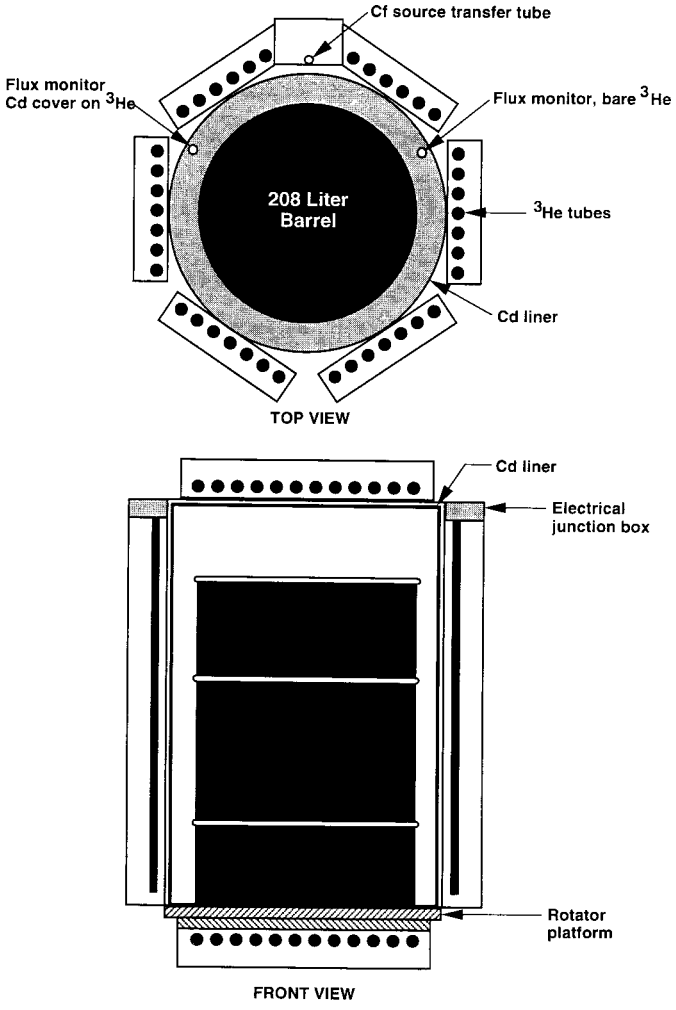
7.2.1 The neutron detectors are ^{3}He-filled cylindrical proportional counters embedded in polyethylene, located around the item in a near 4π geometry. The detection efficiency for neutrons of fission energy should be above about 15 %. Larger detection efficiencies generally provide better precision and lower detection limits for a given count time subject to cycle time, source coupling and other operational parameters. The counter detection efficiency should vary less than 10 % over the item volume with no item present.

7.2.2 The flux monitors are ^{3}He-filled proportional counters mounted on the inner walls of the measurement chamber and not embedded in polyethylene. One flux monitor is covered with Cd approximately 1 mm thick; the other is bare and



NOTE 1—A sketch of a shuffler designed to assay 208-L drums. The source storage shield is a 2000-kg, 1.2-m cube that resides close to the measurement chamber. In this design it is on top of the measurement chamber. This configuration reduces the footprint of the instrument and may reduce the cosmic ray induced background somewhat. Other configurations are also in common use. The stepping motor drives the Cf source through the source transfer (or guide) tube between the storage position and the irradiation position inside the measurement chamber.

FIG. 2 Shuffler for 208-L Drums of Waste



SHUFFLER DETECTOR BANK DIAGRAM

NOTE 1—The front and top views of the measurement chamber shown in Fig. 2 are shown here in greater detail. The 208 L drum sits on a rotating platform above the bottom detector bank. Six side banks surround the item, with the Cf source transfer tube at the back. The two flux monitors are placed at the rear of the item chamber.

FIG. 3 Shuffler Detector Bank Diagram

responds predominantly to thermal neutrons. The Cd shields the so-called fast flux monitor from thermal neutrons; therefore, the two flux monitors can be compared in order to provide information about the neutron energy distribution emerging from the item when the Cf shuffler is brought up. Measured matrix corrections are functions of the fast and thermal flux monitor rates.

7.3 *Shielding*—The quantity of radiation shielding for the ^{252}Cf source is governed by personnel safety requirements although control of the background is also a consideration.

7.3.1 The measurement chamber is typically surrounded by 0.3 to 0.6 m of materials such as polyethylene and borated polyethylene to shield the operator during the ^{252}Cf irradiation.

7.3.2 The shield for the ^{252}Cf storage position is typically about 0.6 m thick (1.2-m cube), depending on the source strength, or the source is placed 1.8 m underground. Composite shields are more effective than polyethylene alone for large ^{252}Cf sources (**29**). The source home position may have a heavy-metal shield to reduce direct gamma dose. The composite shield concept should also takes into account secondary capture gamma-ray generation. If the source store is not directly mated to the measurement chamber, care should be taken in the routing of and shielding to the intervening guide tube so as to manage the time averaged dose rate in the vicinity.

7.4 *Electronics*—High count rate, commercially available nuclear electronics provide standard logic pulses from the ^{3}He-filled proportional counters. These pulses are typically processed by shift register coincidence electronics for the passive measurement, and by gated fast scalers or a multi-channel scaling system for the active measurement. Other correlated neutron counting electronics can be used, with appropriate changes to the data reduction equations.

7.5 ^{252}*Cf Source Drive System*—The source is attached to a flexible drive cable that runs inside a guide tube. The source movement is controlled by stepping motors or an alternative method that offers precise timing, positioning, and computer control. During the active measurement, variations in the timing of the source transit, irradiation or counting portions of the shuffles cause variations in the measured response. Components should be selected to reduce this potential problem to negligible levels.

7.6 ^{252}Cf sources are commercially available and are usually replaced every few years (typically of the order of two half-lifes) subject to preserving desired active detection limits and precisions. The vendor should understand the safety issues and provide guidance in addressing them.

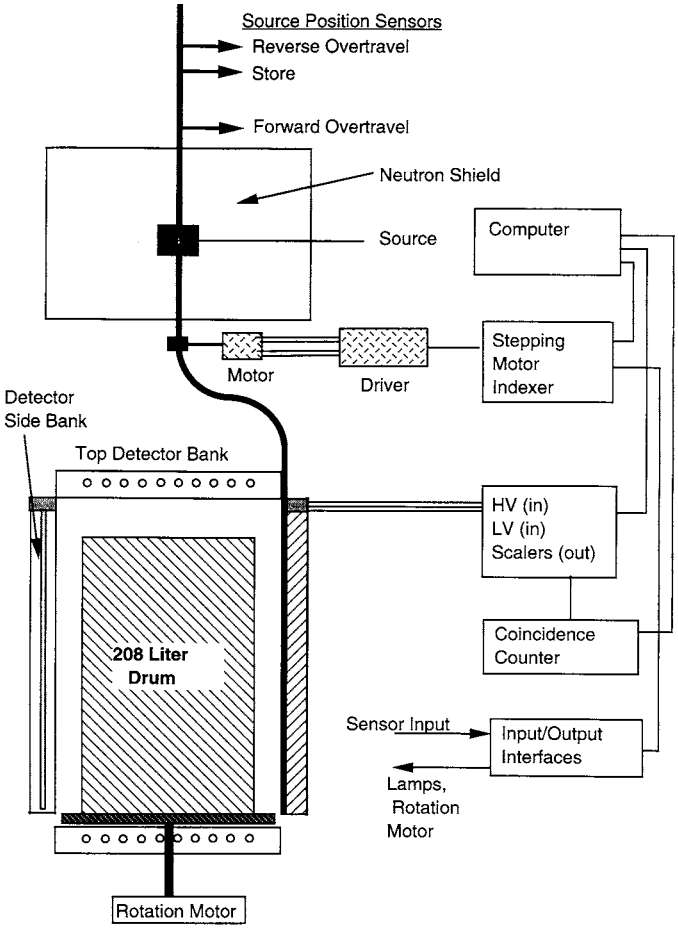
7.6.1 The source vendor should encapsulate the ^{252}Cf, securely attach the source drive cable, provide shielded shipping casks, and assist with the source installation and disposal.

7.6.2 The source vendor should be requested to provide documentation for the ruggedness and integrity of the source encapsulation and perform swipes to demonstrate that the outside of the source capsule is not contaminated.

7.7 Data acquisition and reduction, control of the source motion, and the diagnostic tests require interfacing the instrument to a computer as illustrated in Fig. 4. The computer and software normally are provided by the instrument vendor.

7.8 *Customized Design Issues*:

7.8.1 An initial ^{252}Cf source size of 550 µg is generally adequate for measurements of 208 L drums. Performance for a given source strength can be tailored to some considerable extent by adjusting the chamber design—in particular detection efficiency and source coupling play important roles.



Shuffler Electronic Controls Diagram

NOTE 1—The electrical components and their connections are indicated. The Cf source is moved by the stepping motor and associated driver. Three source sensors are used to verify the source position. The detector signals are amplified and discriminated in junction boxes into which the ^{3}He-filled cylindrical proportional counters are fastened. The logic outputs of the discriminators are fed to scalers and a coincidence counting module. The computer controls the source and rotator and receives the results from the scalers and coincidence counter according to the strict timing sequence in use.

FIG. 4 Shuffler Electronic Controls Diagram

7.8.2 It is recommended that the size of the measurement chamber be just slightly larger than the size of the items to be measured. If small items require measurement in a large measurement chamber, the items should generally be centered in the chamber. Coupling of the interrogation source to the item and of the item to the flux monitors may need special consideration and a container specific calibration will generally be needed.

7.8.3 During an active measurement of a large item, the item should be rotated and the Cf source should scan the vertical length of the item. Some designs use continuous rotation and scanning motion (**2**) while others acquire data using a series of discrete angular and source positions (**21**, **27**, **28**). Discrete scans can provide input for optional analysis algorithms (such as might provide coarse spatial corrections) or might be useful where a symmetric pattern of ^{3}He proportional counters can not be used (for example if the instrument is constrained by the interface to a hot cell).

7.8.4 The standard shuffler configuration assumes some hydrogenous and some metallic matrices will be measured. The interrogation-neutron energies are therefore kept high by not using spectrum tailoring materials between the Cf source and the item being measured and by using a steel reflector behind the Cf source (**1**, **2**). This configuration also includes lining the assay chamber with Cd, which prevents neutrons that are thermalized in the polyethylene of the detector banks from entering the measurement chamber. Thermal neutrons generally penetrate less deeply into the matrix and consequently spatial uncertainties will generally be higher if the matrix and special nuclear material distribution are not homogeneous. Thermal neutrons also are less pentrating into aggregates of special nuclear material. The down side of using a Cd liner, however, is that the sensitivity be over an order of magnitude poorer. The prospects and potential benefits of spectrum tailoring are discussed in (**30**). It should also be noted that some containers (for example, those with concrete liner or known to possess a particular waste characteristics) and some chambers (for example, those requiring significant Pb shielding to control the gamma-ray does rate on the ^{3}He proportional counters) introduce neutron transport peculiarities that should be considered as an integral part of the design process (**21**, **26**, **27**).

7.8.4.1 When it is assured that (*a*) lumps are not a significant problem and (*b*) the matrix is a weak moderator, a polyethylene sleeve can be placed around the assay item for the active mode measurement to reduce the energies of the interrogating neutrons, enhancing the fission rate, the precision, and the sensitivity. A different calibration is necessary for polyethylene "sleeve" measurements. An alternative scheme is to make the Cd liner removable to achieve the same objective (**30**).

8. Hazards

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

8.1.1 Take precautions to maintain personnel radiation exposures as low as reasonably achievable (ALARA). See also Guide C 1592. Typical doses at the surface of the instrument are <20 µSv h^{-1}.

8.1.1.1 The radiation dose from 550 μg ^{252}Cf (unshielded) is about 10 mSv h^{-1} at 1 m, consisting of both gamma and neutron radiation. Large ^{252}Cf sources require remote handling, shielding, and interlocks on automatic transfer mechanisms to help prevent inadvertent or excessive exposure.

8.1.1.2 For large source shields, the gamma rays resulting from neutron capture in hydrogen can contribute significantly to the dose on the outside of the shield; shields loaded with B or Li can greatly reduce this effect.

8.1.2 Take precautions to prevent inhalation, ingestion, or the spread of radioactive contamination. Periodic alpha monitoring of calibration materials, measurement control items, and scrap and waste containers to verify their integrity is recommended. Periodic inspection and monitoring of the shuffler source and guide tube should be carried out.

8.1.3 Take precautions regarding nuclear criticality, especially of unknown items. The measurement chamber approximates a reflecting geometry for fast neutrons. Do not assume that waste is not of criticality concern.

8.1.4 Take precautions to prevent inhalation, ingestion, or the spread of Cd and Pb, if used as shielding. They should be covered with nontoxic materials.

8.1.5 Take precautions to avoid contact with high voltage. The proportional counters require low current supplies of approximately 2 kV.

8.2 The results of this test method might be used to make decisions regarding, for example, the handling and disposal of items or the cessation of safeguards on the items. Consult qualified professionals and Guide C 1490 as needed.

9. Initial Preparation of Apparatus

9.1 The initial preparation of the shuffler passive/active neutron (PAN) apparatus is outlined in 9.2 through 9.6, which discuss the initial setup, calibration, and the initialization of measurement control. The details of preparation are site-specific, dependent on the material categories to be measured, and are generally performed by experts **(31)**.

9.2 *Initial Setup*:

9.2.1 The apparatus weight exceeds 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 shuffler's shielding typically screens the measurement chamber from most sources of background although ultimately detection limits are governed by background conditions **(18, 20)**.

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

9.2.3.1 Most electronics settings are optimized by the manufacturer, and changing them may affect the instrument's performance.

9.2.3.2 The initial setup might include verifying or testing the following items: (*a*) that all software is loaded and running; (*b*) the safety features for the Cf source drive mechanism; (*c*) the operation of the source drive mechanism; (*d*) the status lamps; (*e*) the deadtime coefficients and the coincidence gate length; (*f*) the rotation motor; (*g*) the Cf source transfer velocity, acceleration, and scanning parameters; (*h*) the parallel port inputs and outputs; and (*i*) testing the neutron detection electronics with background and with small sources.

9.3 *Calibration: Preparation*—Use this test method with a scrap and waste management plan that segregates materials with respect to their neutron moderation and absorption properties. References **(2)** and **(32)** describe calibration exercises and provide illustrative data. The passive calibration is conventional (see C 1207) and ^{252}Cf may be used as a surrogate for $^{240}Pu_{eff}$ **(33)**. Additional sources of information can be found in Guides C 1009, C 1068, C 1128, C 1156, C 1210, and C 1215; ANSI Guide 15.20; NRC Guides 5.11 and 5.53; DOE Order 435.1; and U.S. Regulations 10CFR61 and 40CFR141.

9.3.1 Determine the different material types that represent the scrap or waste streams to be measured.

9.3.2 Prepare and characterize the calibration materials. They should represent the material types with respect to parameters that affect the measurement, such as moderation and absorption. The calibration materials should span the special nuclear material mass ranges expected in the scrap or waste to be measured. The fabrication should document traceability for the special nuclear material parameters.

9.3.3 Record the calibration procedure and data. The data should demonstrate the variation of the volume weighted average instrument response as a function of the nuclear material mass and the matrix.

9.3.4 The volume weighted average (VWA) response is an estimate of the count rate that would be obtained from a item containing a homogeneous matrix with a uniform distribution of special nuclear material. One possible way of estimating the VWA response **(2, 34)** is a weighted average calculated from a series of measurements. One or more physically small capsules of special nuclear material of known and ideally low self-shielding are placed in containers filled with uncontaminated matrix material to estimate the response of the instrument to different matrices. Placement is typically along tubes which run the length of the containers and are placed in the matrix at the areal center of equal area columns. For 208 L drums typically 3 to 5 radial positions and 5 to 7 axial positions would be used to define the centroids of the voxels, depending on the severity of the matrix, which defines the spatial gradients. The VWA of the measured response map is computed along with the corresponding standard deviation which is indicative of the potential bias from measurements made with nonuniform (single point-like) distributions of special nuclear material. Spatial mapping using encapsulated sources is also often a pragmatic way to decrease the cost of generating a broad range calibration compared to characterizing and storing suitable distributed calibration materials for large sets of diverse matrices. Spatial maps also lend themselves to numerical spatial integration schemes. Monte Carlo simulations benchmarked to a reference measurement may also be used to generate VWA responses using basic knowledge of the neutron transport properties along with knowledge of the matrix compositions (for example, the measured response at only a single position within a test matrix can be scaled by the calculated VWA-to-point ratio). In this way fewer experimental points are needed which can accelerate the calibration process. As a general rule however, measurements across a set

of test matrices should be made and this is especially useful in establishing flux monitor (or Add-A-Source) trends with matrix characteristics which are more difficult to model accurately.

9.4 *Calibration: Response vs. Mass*—This calibration determines the relationship between the measured instrument response and the mass of nuclear material. If the matrix-specific calibration approach is being used, this calibration data is obtained using the specific matrix found in the unknowns (**32**). Otherwise, a benign matrix is used. The flux monitor data may be recorded for later use in assessing whether the correct matrix-specific calibration is being used. If the polyethylene sleeve is used for measurements of a certain material category, then the calibration data must be acquired with it also (**2**, **32**, **35**).

9.4.1 *active mode*—relates the delayed neutron count rate to the effective or equivalent ^{235}U mass (**7**).

9.4.2 *passive mode*—relates the coincident neutron count rate to the effective mass of ^{240}Pu (**7**).

9.4.3 Determine the range of the calibration. This is often defined by the smallest and largest masses used in the calibration.

9.4.3.1 The best fit to the calibration function within the calibration range sometimes yields nonsensical results outside of the calibration range. Any use of the instrument outside of the calibration range should be evaluated carefully.

9.4.3.2 If the calibration is extended to very small masses, the range should begin at zero instead of the lowest mass used in the calibration. The user should evaluate the response of the instrument with matrix items that contain no special nuclear material.

9.4.4 Measure each calibration mass such that the measurement precision is better than that expected for assay items of similar mass by using longer count times or replicate counts.

9.4.4.1 Measurements of small mass items can have large uncertainties due to lack of signal. If the measurement precision is 10 % or worse, such measurements might be more useful to check the calibration rather than determine it.

9.4.5 Analyze the calibration data to determine an appropriate function. The choice of calibration function will depend on the characteristics of the material categories and the calibration mass range (**1**, **2**, **29**, **32**, **36-43**).

9.4.5.1 Calibration data for waste measurements with small amounts of special nuclear material can generally be fitted with a linear function.

9.4.5.2 If the calibration is extended to very small masses, the calibration might produce less bias if the fit is forced through the origin. The user should verify the appropriateness of this with measurements of matrix material without special nuclear material present.

9.4.5.3 Calibration data for scrap measurements of high mass items may not be suitable for fitting with a linear function.

9.5 *Determining the Matrix Correction*—This section is not applicable if the matrix-specific calibration is being used. It describes a procedure that determines the relationship between the measured flux monitor response and the neutron moderation and absorption effects of the matrix on the measured count rate for uniform items. This relationship will determine a correction to the count rate data that is made before the calibration described in 9.4 is used. Different corrections are required for the active and passive modes.

9.5.1 Determine the range of matrix correction for the active and passive modes separately.

9.5.1.1 At some point, the moderator and absorber content will be sufficiently large as to shield the innermost locations in the item. The user should not try to make a correction for this measurement situation, where special nuclear material could be in the item but not respond.

9.5.1.2 The user must choose how large a response variation with position is acceptable to meet the measurement objectives. A hydrogen density of 0.03 g mL^{-1} will yield a maximum-to-minimum response variation of approximately 2.4 for 208-L drums (**2**).

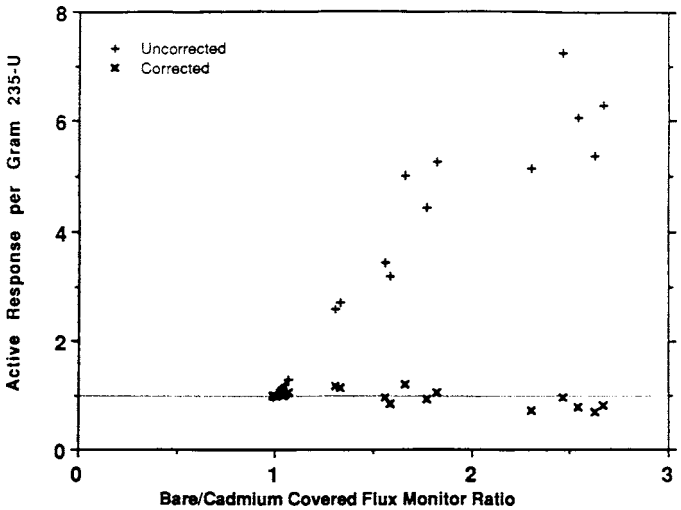
9.5.2 Measure the flux monitor responses and the count rates from the source for each matrix. The measurement precisions should be smaller than those typically obtained in measurements of unknowns or small enough to make an acceptable contribution to the overall measurement error.

9.5.3 Demonstrate that the flux monitor response is adequately independent of the special nuclear material source size and location in the item.

9.5.4 Analyze the data to determine a suitable flux monitor correction function. The choice of correction function will depend on the characteristics of the material categories. Several functions have been used to perform an empirical fit to this type of data (**2**, **12**, **17**, **29**, **38**).

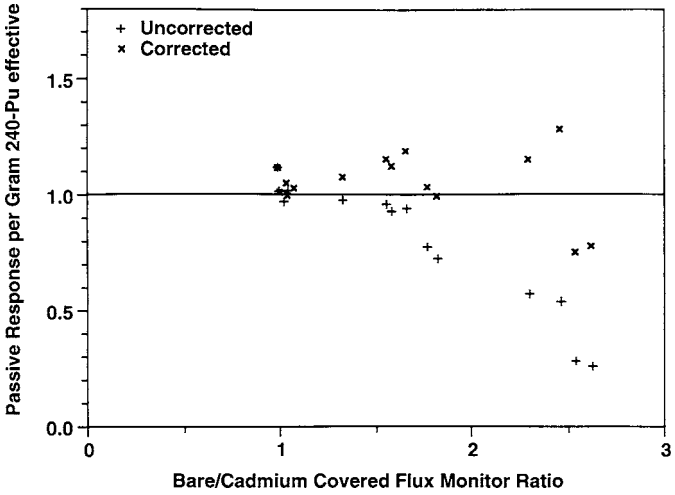
9.5.4.1 The corrected data in Fig. 5 for passive and Fig. 6 for active measurements of homogenous distributions of ^{235}U, shown only as an example, both used the following empirical functional form (**2**):

$$CF = 1/R^{p(R)}, \text{ where the exponent } p(R) = a1\,R^2 + a2\,R + a3 \quad (1)$$



NOTE 1—The measured active response per gram of ^{235}U in 208-L drums is shown for 20 matrices. Both the uncorrected response (+) and the flux monitor corrected response (x) are plotted. The relative standard deviation of the corrected responses is 14 %. The matrices span a wide range of characteristics typical of those found in facilities (**2**). The largest hydrogen content in a matrix was 9.65 kg; the largest boron content was 0.20 kg.

FIG. 5 Active Response as a Function of Flux Monitor Ratio



NOTE 1—The measured passive response per gram of $^{240}Pu_{eff}$ in 208-L drums is shown for 18 matrices. Both the uncorrected response (+) and the flux monitor corrected response (x) are plotted. The relative standard deviation of the corrected responses is 12 %. The matrices cover a wide range of characteristics typical of those found in facilities (**2**). The largest hydrogen content in a matrix was 9.65 kg; the largest boron content was 0.20 kg.

FIG. 6 Passive Response as a Function of Flux Monitor Ratio

where:

CF	=	the rate correction factor. In Section 11 we use subscripts a and p to indicate the active and passive correction factors respectively,
R	=	bare-to-Cd-covered flux monitor response ratio,
a1, a2 and a3	=	fitted coefficients specific to the mode (passive or active) and instrument.

9.5.5 An alternative approach is the matrix-specific calibration, where the user attempts to match the matrix effects of the unknown items with the calibration items (**32**). This approach might use the flux monitor data to verify that the calibration and item matrices are suitably matched.

9.6 *Initialize Measurement Control*—The need for adjustment of the instrument can be determined by measurement control procedures (**44**) (ANSI N15.36). These procedures make use of background measurements, replicate measurements of a specific item, and periodic remeasurement of certain items.

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

9.6.2 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.

9.6.3 Documentation of the measurement control of the instrument may be required (that is, DOE Order 474.1).

9.6.4 The choice of control limits and the action required after a "failure" should take into consideration the measurement uncertainties and the probability of a false positive (**44**).

10. Procedure

10.1 After calibration, the procedure consists of measurements of items with unknown special nuclear material content and measurements that demonstrate that the apparatus is calibrated and functioning properly (measurement control).

10.2 *Measurement Control*—Measurement control measurements are made before assays of unknowns and are interspersed between measurements of unknowns to verify proper functioning of the instrument. If the measurement control indicates the instrument response has changed, determine the cause and make the necessary repairs. In addition, all measurements of unknowns since the last successful test are suspect and may need to be repeated.

10.2.1 *Background Measurements*—Perform periodic background measurements (**44**).

10.2.1.1 *Passive Mode*—Traditional practice is to perform these measurements daily with no special nuclear material in the assay chamber. Low total neutron count rates verify that no breakdown of the proportional counters or their electronics has occurred. Count rates of zero suggest the detector high voltage is off, part of the detection electronics is nonfunctional, or the detector electronics are disconnected. This background measurement is generally used in the passive calculations.

10.2.1.2 *Active Mode*—A background measurement is made at the start of each assay while the item is in the assay chamber, before the source shuffles begin. For a combined PAN assay the active background is usually the non-deadtime corrected passive data.

10.2.2 *Measurement Control Bias Measurement*—Perform periodic measurements of stable items containing special nuclear material to verify the stability of the instrument response (**44**). Typically high and low masses are used on different days. Traditional practice is to perform a daily measurement for instruments used daily although more frequent state of health checks may be made subject to an application specific consequence analysis. For instruments used intermittently, this check is recommended before and after each use. Agreement with the previous value within the control limits indicates long-term stability of the instrument's response. Long-term stability suggests that the calibration is still valid. Low results may indicate that a detector or detector bank is not functioning correctly. High results may indicate electrical noise.

10.2.2.1 The measurement control item used for the check must provide a consistent response. Corrections should be made for radioactive decay.

10.2.2.2 The uncertainty estimated from counting statistics for these measurements will be constant for a given count time, except for changes due to source decay. Otherwise, the source of variation should be investigated.

10.2.3 *Measurement Control Precision Measurement*—Perform periodic replicate counts of different items to verify the estimates of the measurement precision (**44**). This test might be conducted monthly or after each calibration. Statistical agreement between the standard deviation of the replicates and the uncertainty estimate from a single measurement's

counting statistics indicates short-term stability of the instrument's response. Lack of agreement might indicate background variations, electrical instabilities, mechanical changes, or errors in the implementation of the software algorithms.

10.3 *Item Measurements*:

10.3.1 Position the item to be measured in the counting chamber. The counting geometry should be the same for all measurements. If the polyethylene sleeve is used for assay of an item then the calibration used for the analysis should have been obtained in the "sleeve" configuration.

10.3.2 Measure for the chosen count times. It is often advisable to measure unknowns and measurement control items for the same count times so as to eliminate this as a potential source of error.

10.3.2.1 *Passive Mode*— The passive count time is typically between 400 and 1000 s. When a matrix correction is desired, the passive count is followed by a short count (on the order of 10 to 100 s) with the ^{252}Cf source interrogating the item in order to gather the necessary flux monitor rates. Additional useful information might also be obtained at this time (see Section 10.3.4.3). The drum may be rotated an integral number of times during the flux monitor determination. Other experimental passive matrix correction techniques may optionally be incorporated and used (for example, the Add-A-Source method **(36)**).

10.3.2.2 *Active Mode*—For a shuffler of the type illustrated in Fig. 3 and Fig. 4 the item is usually rotated during the active measurement, asynchronously with the ^{252}Cf source motion. The active count (for a total assay sequence of about 1000 s) generally consists of a 250-s background count of the item with the shuffler source stored, followed by approximately 30 shuffles of the ^{252}Cf source, each with an interrogation of about 10 s and a delayed neutron count time of about 10 s. One-way source transit times are less than about 1 s. The cumulative delayed neutron counting period would be $30 \times 10 = 300$ s in this example.

10.3.3 When the counts are complete, document the measured quantities.

10.3.3.1 *Passive Mode*—Compute the deadtime and background corrected totals, reals and flux monitor rates along with their associated precison. (See Test Method C 1207 for additional details on coincidence counting).

10.3.3.2 *Active Mode*—Compute the deadtime and background corrected delayed neutron rate.

10.3.4 The following diagnostic tests are recommended for each measurement.

10.3.4.1 *Passive Mode*—(*a*) The total neutron count rate can be used to estimate the accidentals rate **(4, 41)**. Lack of agreement within statistical uncertainties between the estimated and measured accidentals count rates suggests a hardware failure in the coincidence circuitry or that the background neutron count rate changed significantly during the measurement. Note that for a symmetric counter and fairly homogeneous items the passive rate should remain approximately constant as the item is rotated. For some designs, however, the item must be held fixed during data acquisition and indexed to obtain a rotational average for this test to pass. (*b*) Each measurement can be divided into several short counting periods, and statistical tests performed looking for outliers in the individual counting periods **(12, 36, 41, 45, 46)**. This "outlier" test reduces the effects of cosmic ray background or of changing conditions during the measurement. Outliers are generally replaced with data from an additional counting period, which is obtained without operator intervention by the software.

10.3.4.2 *Active Mode*—(*a*) A detector bank with zero counts is suspect and reported with an error message **(1,2)**. This error condition might indicate the detector bank is not functional. If backgrounds are very low in every detector bank, this diagnostic might be more confusing than helpful; (*b*) Ratios of counts in different detector banks can be compared with historical values; if a ratio is statistically out of bounds, an error message can be generated **(1,2)**. This error condition might indicate that either a detector bank is not functioning correctly or the assay item is not suitable for measurement. For low count rates, the value of this diagnostic is also low; (*c*) The overall regularity of the various phases of an assay can be checked by calculating a quantity **(1,2)** from the measured times for motion of the ^{252}Cf source and the count times. This quantity is compared to the value calculated using the nominal times for motion and counting. If the two values differ by more than expected, a hardware failure in the source motion controller or the clock might be suspected.

10.3.4.3 *Active Mode*—The neutron transmission through the item has been used to evaluate whether the item behaves similarly to the calibration items **(32)**. During an irradiation with the ^{252}Cf source, compare the measured count rate in the opposing detector banks with the rates obtained with the calibration items. A statistically significant difference suggests that the wrong calibration is being used. A very low value suggests that inadequate penetration of the item has occurred, the measurement is not sensitive to the center of the item, and the potential exists for undetected nuclear material to be in the center of the item. It is possible to use the flux monitor count rates in a similar manner **(2)**.

10.3.5 Calculate the amount of special nuclear material (for example, U, Pu or both) in the item.

10.3.6 If replicate measurements are performed, wait at least four minutes after the ^{252}Cf irradiation ends before starting the next assay to allow the induced delayed neutron signal to decay to negligible levels.

10.3.7 Remove the item from the counting chamber.

11. Calculations

11.1 This section provides a summary of the calculations developed for a 208-L system. Use of other electronics may require different equations. The calculations are usually performed by the software, not by the user. The vendor should provide quality assurance that the calculations are correctly implemented in the software. The calculations follow the same general approach whether the results are used for calibration, measurement control, or determining an unknown. Estimates of the measurement uncertainties follow standard propagation techniques and are detailed in the references.

11.2 *Passive Assay for Plutonium*—Data reduction for the passive coincidence measurement is explained in Test Method C 1207 and (**2, 4, 41, 46**). Most of the variables were defined in 10.2.3.

11.2.1 Determine the total neutron count rate and its random statistical uncertainty.

11.2.2 Determine the coincidence count rate and its random statistical uncertainty. There are approximations for statistical uncertainty (see (**4, 12**) and Test Method C 1207) which use the cumulative shift register results, however, if the count has been broken into a series of shorter interval the scatter in the data can be used directly.

11.2.3 Correct the count rates, including those for the flux monitors, for deadtime (**4, 29**).

11.2.4 Perform a background subtraction.

11.2.5 *Matrix Correction*:

11.2.5.1 If the matrix correction calibration is being used, make a correction to the deadtime and background corrected reals rate for matrix effects using the flux monitor responses (**1,2, 12, 36**) obtained during the ^{252}Cf interrogation.

$$R_c = R'' \times CF_p(\text{flux monitor}) \quad (2)$$

where:

the passive flux monitor correction, CF_p (flux-monitor), is discussed in 9.4 and (**2**).

11.2.5.2 If the matrix-specific calibration approach is being used, no matrix correction is made.

11.2.6 As the assay gets larger, effects that have the potential to cause bias, like multiplication, become more important, if not corrected (**4, 41, 42, 46**).

11.2.7 Use the calibration function, the parameters described in 9.3, and R_c to calculate the assay result in terms of the ^{240}Pu$_{\text{eff}}$ mass, m_{eff}. The ^{240}Pu$_{\text{eff}}$ mass is defined in terms of the masses, m_A, of the Pu isotopes, A, as:

$$m_{eff} = 2.52\, m_{238} + m_{240} + 1.68\, m_{242} \quad (3)$$

where the numerical weighting factors are nominal nuclear data values. From time to time revised nuclear data evaluations become available and the coefficients may also be slightly dependent on chamber design. For many situations, ^{238}Pu and ^{242}Pu are minor contributors and the results are insensitive to the exact values provided the same values are used for calibration and for the analysis of the assay of unknown items.

11.2.7.1 Waste measurement data are generally suitable for direct proportionality between R_c and m_{eff}.

11.2.7.2 Scrap measurement data might require a general quadratic function (**41, 42, 46**):

$$R_c = k0 + k1\, m_{eff} + k2\, (m_{eff})^2 \quad (4)$$

where:

$k0$, $k1$, and $k2$ are the calibration constants determined in 9.3.

11.2.7.3 Other methods of data analysis are available that take advantage of additional information when it is available (**4, 41, 42, 46**).

11.2.8 Use the isotopic ratios and m_{eff} to calculate the assay result in terms of total Pu:

$$\text{mass}_{Pu} = m_{eff}/(2.52\, f_{238} + f_{240} + 1.68\, f_{242}) \quad (5)$$

where:

f_A = the weight fraction of Pu isotope A.

11.2.9 The estimate of the measurement uncertainty should include the components that cause significant effects. These generally include counting statistics, calibration errors, uncertainties in the matrix correction factor, and uncertainties in the isotopic ratios. Some components may be difficult to quantify.

11.3 *Active Assay for Uranium*—Data reduction for the active shuffler measurement is explained in (**1, 2**) . Most of the variables were defined in 10.2.3.

11.3.1 The uncorrected delayed neutron count rate, *ucr*, is computed from the total counts and time.

$$ucr = T1/t1 \quad (6)$$

where:

$T1$ = total neutron counts acquired over the n shuffles, and

$t1$ = total counting time for the shuffles evaluated from the product of the number of cycles and the delayed neutron counting period per cycle.

It is assumed that the same number of shuffles and the same time pattern are used for unknowns and calibration measurements, otherwise, make the necessary corrections (**1, 47**).

11.3.2 Subtract the background count rate, derived from the total active background counts, *bkgd*, accumulated:

$$ucr' = ucr - (bkgd/t2) \quad (7)$$

where:

$t2$ = background time.

11.3.3 Make a correction for the radioactive decay of the interrogation source, allowing for the isotopic composition of the Cf in the source if necessary:

$$ucr'' = ucr' \times \exp(\lambda t') \quad (8)$$

where:

λ = the effective decay constant, and

t' = the time since the reference date of the calibration.

11.3.4 Use the same count times, source transfer timing, and number of shuffles for each measurement, or correct for the differences (**1**).

11.3.5 Use the flux monitor matrix correction factor to correct the delayed neutron signal for matrix effects (**1, 2, 29**).

$$ccr = ucr'' \times CF_a(\text{flux monitor}) \quad (9)$$

where:

CF_a(flux monitor) is discussed in 9.4 and (**2**).

11.3.5.1 If only one matrix is being assayed and the appropriate calibration materials were used for it, the user might use the matrix-specific calibration instead of applying the matrix correction factor.

11.3.6 No corrections are made for self-shielding and heterogeneity (**1,2**).

11.3.7 Use the calibration function and parameters to compute the assay result in terms of ^{235}U mass from the corrected measured response, *ccr*.

11.3.7.1 Waste measurements involving U of a single enrichment generally use a linear function:

$$ccr = k_{235}\, m_{235} \quad (10)$$

where:

k_{235} = calibration constant for ^{235}U, and

m_{235} = mass of ^{235}U.

11.3.7.2 If a wide range of U enrichment is to be measured, or if Pu was found in the passive assay, then additional corrections may be required (**1**,**2**, **29**).

11.3.8 Use isotopic information and the ^{235}U mass to get the assay result in terms of U mass.

$$m_{\text{uranium}} = \frac{m_{235}}{f_{235}} \tag{11}$$

where:

f_{235} = weight fraction of ^{235}U.

11.3.9 The estimate of the measurement uncertainty should include the components that cause significant effects. These generally include counting statistics, calibration errors, uncertainties in the matrix correction factor and uncertainties in the isotopic ratios. Other components may be difficult to quantify.

12. Precision and Bias

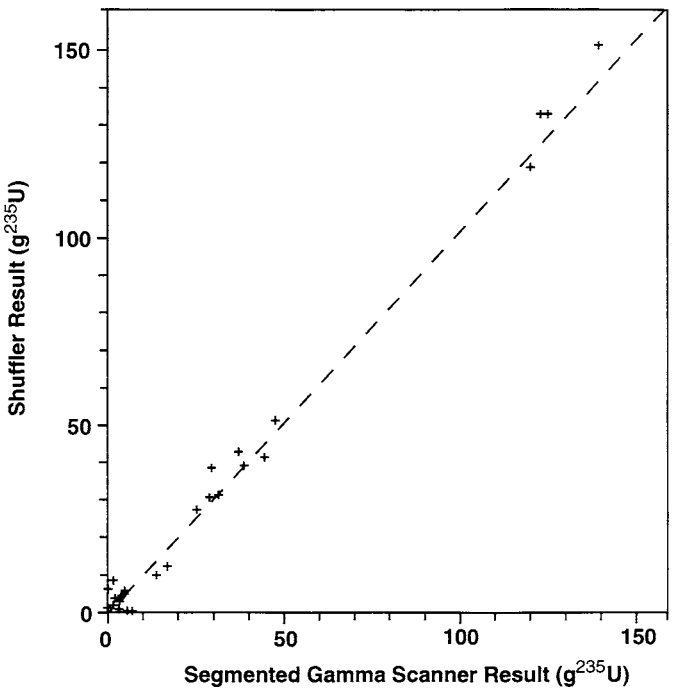
12.1 The precision and bias of shuffler measurements are functions of several interrelated factors, consequently a simple precision or bias statement is not possible (**2**, **34**, **35**). The interrelated factors include facility-specific procedures, matrices, chemical forms, and quantities. This section provides information but cannot substitute for critical thinking, professional skill, and verification measurements. The evaluation of the uncertainty for a shuffler assay is not a purely mathematical task, it requires detailed knowledge of the measurement method, the procedures, and the items being measured. The evaluation should be documented as part of the total measurement uncertainty assessment and reviewed by an NDA profession as defined in Guide C 1490 prior to the instrument entering operation. Measurements of uncharacterized drums generally yield results of indeterminate accuracy. However, a combination of measurement methods applied to such drums may be used to estimate measurement uncertainties. Except for measurements of small quantities of nuclear material, the possibility of bias is of greater concern than the issue of inadequate precision (**2**, **35**). This section lists precision and bias statements applicable to both passive and active measurements, then statements specific to the passive mode, then statements specific to the active mode.

12.2 *Passive and Active Measurements*—Each user of this test method should estimate the precision and bias for each scrap and waste category. See (**2**, **29**, **32**, **35**, **40**) as examples.

12.2.1 A comparison with another assay technique can be helpful to estimate potential bias. Figs. 7-9 are examples of such comparisons. In general, other techniques (for example, Test Method C 1133 or Test Method C 1207) are susceptible to different sources or magnitudes of bias.

12.2.2 Calibration exercises can be helpful to estimate potential bias (**2**). In general, it may be difficult to determine how well the calibration matrices emulate the unknowns.

12.2.3 Destructive analysis is generally not practical as a source of bias information when the items are heterogeneous.



NOTE 1—The results of active shuffler measurements for the ^{235}U content of 208-L drums are compared to the results of segmented gamma scanner (SGS) measurements (**32, 35**). These drums contain low-density waste with unknown U loadings from an enrichment plant located in Portsmouth, Ohio. The matrices are alumina, low-density combustible waste, ABSORBAL, and oil-soaked 3M cloth. The SGS results are from a direct measurement of the drum (if the transmission at 186 keV was possible) or from summing the results of measurements on 127 mm (5-in.) cans whose contents were poured into the drums. The measurement uncertainties for both techniques in this example are dominated by bias rather than precision. The lack of agreement below about 9 g suggests additional information (another measurement technique perhaps) is needed if a better understanding of the bias is desired.

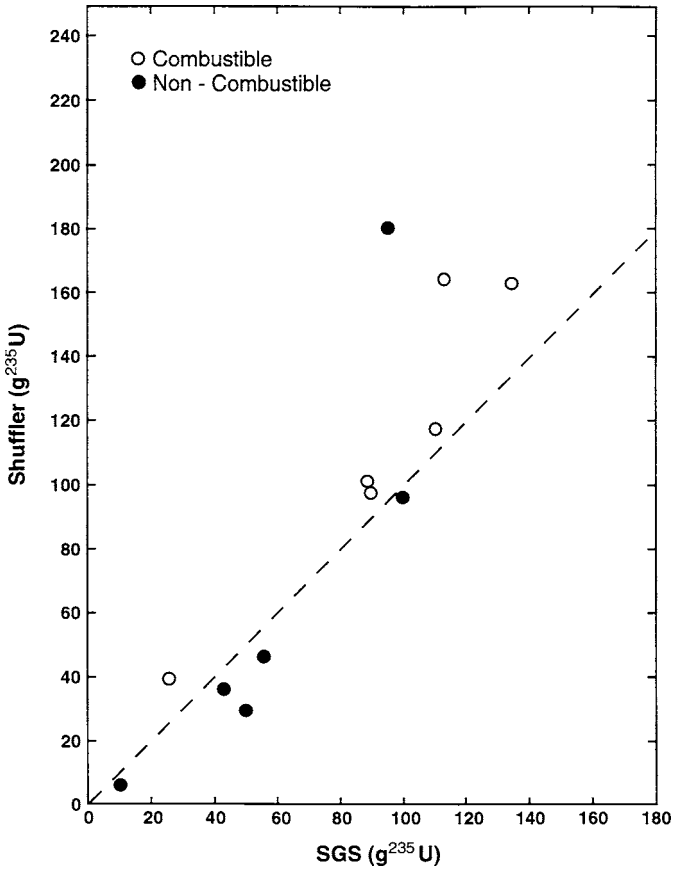
FIG. 7 Comparison Between Active Mode and Segmented Gamma Scanner Results from an Enrichment Facility

12.3 The precision of a shuffler measurement can be estimated using statistical calculations on the data from a single measurement or it can be evaluated by replicate measurements.

12.3.1 Longer counting times (that is, a greater number of shuffler cycles in active mode), more nuclear material, or the use of an apparatus with higher detection efficiency will improve the measurement precision. In general, where throughput and other factors are driving considerations, the counting sequence is chosen to make best use of the assay time available given based on what is known about the measurement scenario.

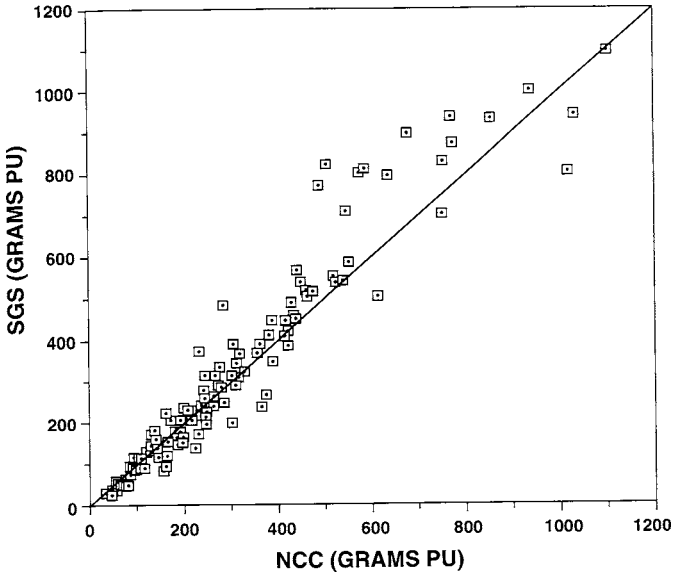
12.4 The bias of a shuffler assay is dependent upon many factors relating to the segregation and packaging of the matrix materials as well as the physical and chemical properties of the nuclear material. If the criteria of 5.4 and 9.2 are not met, the bias is indeterminate.

12.4.1 Biases can occur if the characteristics of the calibration materials differ significantly from those of the item being measured.



NOTE 1—The results of active shuffler measurements for the ^{235}U content of 208-L drums are compared to the results of segmented gamma scanner measurements. These drums contain either low-density combustible waste or medium-density non-combustible waste from a bulk-processing facility for HEU located in Erwin, Tennessee (**46**). The measurement uncertainties for both techniques are dominated, in this example, by bias rather than precision.

FIG. 8 Comparison Between Active Shuffler Mode and Segmented Gamma Scanner Results from a Fuel Fabrication Facility



NOTE 1—The results of passive shuffler mode (Neutron Coincidence Counter, NCC) measurements for the Pu content of 19-L (5 US-gallon) pails are compared to the results of segmented gamma scanner measurements. The pails contain scrap and waste generated by a reprocessing facility in Aiken, South Carolina (**40**). The matrices had no hydrogen. The measurement uncertainties for both techniques are dominated, in this example, by bias rather than precision.

FIG. 9 Comparison Between Passive Shuffler Mode and Segmented Gamma Scanner Results

12.4.2 Mixing material from different matrix segregation categories can lead to a situation where no calibration is appropriate.

12.5 Moderators and absorbers in the matrix can cause bias effects. Over some range these effects can often be adequately corrected; large quantities, however, may bias the result high or low (**2**).

12.5.1 Hydrogen is the element with the largest potential for causing a bias due to moderation and absorption effects. Reference (**2**) describes a method to determine corrections valid for hydrogen levels up to 0.04 g mL^{-1}.

12.6 Bias effects can occur as a result of varying fill heights, heterogeneity, or item positioning because the detection efficiency is not constant over the assay volume. Spatial effects for the detection efficiency vary as much as 10 % for the totals and delayed neutron count rates, and as much as 19 % for the coincidence count rate over the volume of the assay chamber for a benign matrix (**29**).

12.7 Calibration materials have assigned values for mass and isotopic ratios. A bias in an assigned value causes a bias in the calculated results. Where practical, this source of bias should be made negligible in the context of the measurement objectives. In general the uncertainty contribution needs to be quantified and propagated into the final assay result.

12.8 This test method requires knowledge of the isotopic ratios to compute the total element mass from the measured response. A bias in the isotopic ratios will cause a bias in the calculated results.

12.9 Proper adjustment of electrical circuit parameters such as the pre-delay and those controlling the californium source motion can eliminate them as a possible source of significant bias (**1**,**2**).

12.10 If only Pu is present, both the active and passive modes can be used to assay for it, although, subject to the quality of the knowledge of the relative isotopic composition and the counting sequence, the passive result will generally be less biased and have better precision. The active and passive measurements are partially independent and so a lack of agreement between the two results, outside the assigned joint confidence intervals, indicates at least one measurement is biased. Depending on the measurement objectives this may require expert review on a case by case basis.

12.11 *Active Measurements*—Fig. 7 and Fig. 8 compare the results of active mode measurements of 208-L drums with segmented gamma scanner measurements (Test Method C 1133). These items are process waste with unknown ^{235}U content. Fig. 7 compares the shuffler results to segmented gamma scanner results for low-density waste from a U enrichment plant (**32**, **35**). Fig. 8 compares the shuffler results to segmented gamma scanner results for two waste categories

from a naval fuel fabrication plant (**48**). The measurement uncertainties for these results are dominated by bias effects except at very small masses. The difference between the active mode and the segmented gamma scanner results is an estimate of the bias in these measurements.

12.12 Counting statistics typically contribute a random error of <2 % (relative standard deviation) for a 1000-s assay of a 208-L drum containing 5 g of ^{235}U subject to matrix severity. For larger quantities of non-lumped ^{235}U, the random error is generally below 1 % (relative standard deviation) under similar conditions (**2, 29**). To the extent that they can be controlled, precisions fit for purpose should be obtained.

12.12.1 Use of a larger ^{252}Cf source will improve the precision of the active measurement. Flux monitors and shielding are designed with a source strength and item set in mind. Uprating of the source therefore first requires an evaluation of the associated consequences.

12.13 Bias values of approximately 0.2 % have been reported for measurements of kg quantities of ^{235}U in 0.5 L containers of homogeneous product material in 1000-s count times, when compared to destructive analysis (**49**).

12.14 Bias values due to matrix effects of 25 % have been reported for measurements of U and Pu in 208-L mock-waste drums performed during calibrations. A flux monitor correction was applied and 1000-s count times were used. The majority of the data was taken with either 5 g of HEU or 30 g of Pu placed in a variety of matrices (**2, 34**).

12.15 Neutron absorbers such as boron cause effects that can be corrected with the matrix correction factor when the moderator density is low (that is, H density <0.04 g mL^{-1}). Larger quantities of moderator will cause these absorber effects to bias the measurement results (**2**).

12.16 Self-shielding effects for active neutron measurements are minimized when the irradiating neutron energies are kept relatively high and the nuclear material is distributed. Large quantities of moderating material in the matrix are the dominant factor affecting the irradiating neutron energies for large containers.

12.16.1 Calculations predict that a 30-g Pu metal sphere placed in an empty 208 L drum yields 70 % of the response of thirty 1-g spheres distributed uniformly in the drum (**2**). However, with a matrix of polyethylene shavings (estimated to contain a hydrogen density of 0.009 g mL^{-1}), self-shielding causes significant bias for the 1-g sphere of Pu metal (**2**).

12.16.2 Calculations predict that 5 g of HEU diluted in a small capsule (approximately 35 mL) placed in an empty 208 L drum yields 92 % of the response of 5 g distributed uniformly (**2**). This result is consistent with the reported measurements.

12.16.3 Calculations predict that a 10-mg metallic U (93 % enriched) sphere placed in an empty drum yields 86 % of the response of 10 mg distributed uniformly. However, a 100-g sphere of U oxide yields 58 % of the response of 100 g distributed uniformly (**34**).

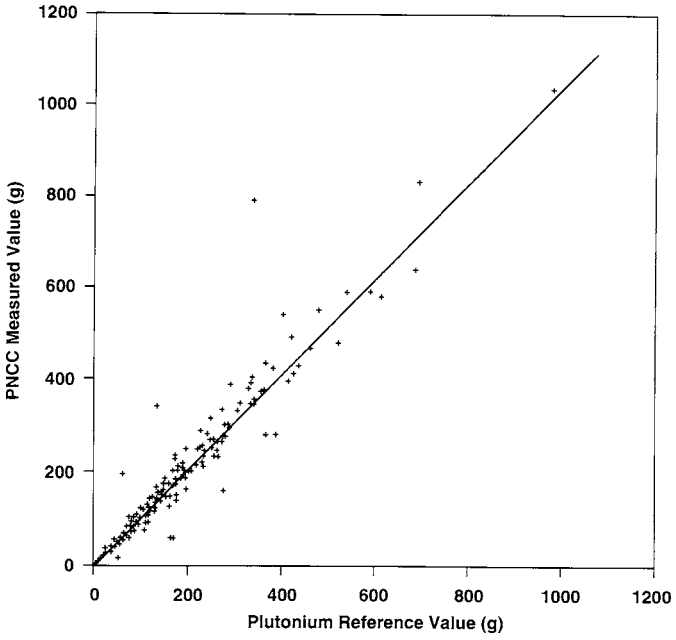
12.17 The active-mode background correction uses a background count at the beginning of each measurement (with the ^{252}Cf source in the storage location). This correction reduces the bias from sources such as (α, n) reactions, the Cf source in its shield, or cosmic rays to negligible levels (**1,2**).

12.18 *Passive Measurements*—Fig. 9 and Fig. 10 compare the results of passive mode measurements, using matrix-specific calibrations, with the results of alternative techniques. Fig. 9 compares passive measurements with segmented gamma scanner measurements (Test Method C 1133) for 19 L (5 US gallon) pails containing Pu scrap (**40**). Fig. 10 compares passive measurements with calorimetric assay. These matrices do not contain hydrogen and were generated at two Pu purification plants. The measurement uncertainties are dominated by bias effects, except for the results at very small masses. The differences between the two techniques are an estimate of the bias in these measurements.

12.19 Counting statistics contribute a random error of less than 1 % for a 1000-s count of an item containing more than 1 g $^{240}Pu_{eff}$ of PuO_2 (**39**). The precision is better for assays of larger quantities of Pu.

12.20 Bias values of 2 % have been reported for measurements on small containers (1-L metal cans) of homogeneous Pu material, when compared to destructive analysis (**39**). The Pu masses ranged from 75 to 874 g; the counting times were 1000 s; and no matrix correction was applied.

12.21 Bias values of 10 % have been reported for measurements of the Pu content in 208-L waste drums, performed during calibration. A flux monitor correction was applied and



NOTE 1—The results of passive shuffler mode (Passive Neutron Coincidence Counting, PNCC) measurements for the Pu content of small containers of scrap and waste are compared to the (reference) results of calorimetry and gamma ray isotopics measurements (CAL/ISO). The containers were generated by a Pu R & D facility in Los Alamos. The matrices do not contain hydrogen. The measurement uncertainties are dominated by bias rather than precision, except at the lowest masses, for this data set. The outliers could be due to transcription errors or a bias in either technique.

FIG. 10 Comparison Between Passive Mode and Calorimetry Results

1000-s count times were used. Most of the data is from 30 g of Pu placed in a variety of matrices (**2, 34**).

12.22 The bias due to cosmic-ray background can be on the order of 0.02-g $^{240}Pu_{eff}$ for large dense items at sea level and can double at an elevation of 2000 m (**4**). Corrections can be made if necessary (**45**). Cosmic ray induced rates depend on the mass and atomic number of the matrix materials, Pb having a far higher specific production rate than most other common matrix materials such as combustibles and steel (**19**).

12.23 The coincidence requirement corrects the passive measurement for the presence of (α, *n*) neutrons. When multiplication is present, a bias can result. The bias effects due to neutron multiplication increase with Pu mass and are affected by variations in the distribution of the Pu and the presence of moderating and (α, *n*) materials.

12.23.1 Waste measurements of matrices with uniformly distributed special nuclear material in 208-L drums exhibit negligible multiplication.

12.23.2 A multiplication effect of 9 % on the neutron coincidence rate has been reported for 10 g of $^{240}Pu_{eff}$ present in Pu oxide (**39, 40**). The effect will decrease if the Pu is diluted by a matrix.

12.24 Neutron absorbers have negligible effects on the accuracy of passive coincidence measurements of unmoderated samples.

12.25 Self-shielding effects do not exist for passive neutron measurements.

13. Keywords

13.1 active neutron measurements; californium; californium-252; NDA; neutron coincidence counting; PAN; passive neutron measurements; plutonium; scrap measurements; shuffler; uranium; waste measurements

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