Designation: C 1316 – 01

Standard Test Method for Nondestructive Assay of Nuclear Material in Scrap and Waste by Passive-Active Neutron Counting Using a ^{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 for uranium and plutonium content using a ^{252}Cf shuffler. Shuffler measurements provide rapid results and can be applied to a variety of matrix materials in containers as large as 208-litre drums. Corrections are made for the effects of matrix material. This test method has been used to assay items containing uranium, plutonium, or both. 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 to measure ^{238}Pu, ^{240}Pu, and ^{242}Pu. It has been used to assay items with plutonium contents between 0.03 g and 1000 g. It could be used to measure other spontaneously fissioning isotopes. 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 californium source and counting of the delayed neutrons from the induced fissions to measure ^{235}U. 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 fissionable isotopes.

1.2 This test method requires knowledge of the relative isotopic composition to determine the mass of the different elements.

1.3 This test method may give biased results for measurements of containers that include large quantities of hydrogen.

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:*

C 859 Terminology Relating to Nuclear Materials[3]

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

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

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

C 1068 Guide for Qualification of Measurement Methods by a Laboratory Within the Nuclear Industry[3]

C 1128 Guide for Preparation of Working Reference Materials for Use in the Analysis of Nuclear Fuel Cycle Materials[3]

C 1133 Test Method for Nondestructive Assay of Special Nuclear Material in Low Density Scrap and Waste by Segmented Passive Gamma-Ray Scanning[3]

C 1156 Guide for Establishing Calibration for a Measurement Method Used to Analyze Nuclear Fuel Cycle Materials[3]

C 1207 Test Method for Nondestructive Assay of Plutonium in Scrap and Waste by Passive Neutron Coincidence Counting[3]

C 1210 Guide for Establishing a Measurement System Quality Control Program for Analytical Chemistry Laboratories Within the Nuclear Industry[3]

C 1215 Guide for Preparing and Interpreting Precision and Bias Statements in Test Method Standards used in the Nuclear Industry[3]

2.2 *ANSI Documents:*

[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 Nondestructive Assay.

Current edition approved June 10, 2001. Published September 2001. Originally published as C 1316 – 95. Last previous edition C 1316 – 95.

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

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

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 859.

3.2 *Definitions of Terms Specific to This Standard:*

3.2.1 *accidentals (a)*, *n*—the number of neutrons detected in the (*a*) gate interval following the initial detection of each neutron during the selected count time, *t*. These neutrons come from many sources and are not physically correlated with the initial neutron.

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

3.2.3 *benign matrix*, *n*—a matrix that has negligible effects on the neutron transport. A benign matrix includes very little neutron moderator or neutron absorber.

3.2.4 *coincidence gate length*, *n*—the time interval following the detection of a neutron during which additional neutrons are considered to be in coincidence with the original neutron.

3.2.5 *coincident neutrons*, *n*—neutrons emitted simultaneously from a single event. Two or more coincident neutrons are correlated in time with the occurrence of one event, such as fissioning of a nucleus.

3.2.6 *die-away time*, *n*—the average lifetime of a neutron from the time of emission until the neutron is detected. The average lifetime is the time required for the neutron population to drop to 1/e of the original value. Die-away time is a function of several parameters including the detector design, the assay item characteristics, and the neutron energies.

3.2.7 *effective ^{240}Pu mass (m_{eff})*, *n*—the mass of ^{240}Pu that would produce the same coincidence response in the instrument as the assay item. It is a function of the quantities of the even-mass isotopes of plutonium and fundamental nuclear constants. It is specific to the type of coincidence circuitry used **(4)**.

3.2.8 *flux monitors*, *n*—detectors in the measurement chamber that measure the interrogating neutron flux.

3.2.9 *item*, *n*—the entire scrap or waste container being measured and its contents.

3.2.10 *lump*, *n*—that contiguous mass of nuclear material that is sufficient to affect the measured signal.

3.2.11 *lumps*, *n—in the context of the active measurement mode*, have a dimension larger than the mean free path of an interrogating neutron and consequently exhibit self-shielding.

3.2.12 *lumps*, *n—in the context of the passive measurement mode*, have a dimension larger than the mean free path of a fission neutron and consequently exhibit multiplication.

3.2.13 *matrix*, *n*—the material that comprises the bulk of the item, except for the special nuclear material and the container. This is the material in which the special nuclear material is embedded.

3.2.14 *matrix-specific calibration*, *n*—uses a calibration matrix similar to the waste matrix to be measured. No matrix correction factors are used; this calibration is generally not appropriate for other matrices.

3.2.15 *neutron absorbers*, *n*—materials that have relatively large absorption cross sections for thermal neutrons. Absorbers with the largest cross sections are commonly known as neutron poisons. Some examples are lithium, boron, cadmium, and gadolinium.

3.2.16 *neutron coincidence counting*, *n*—a technique used to measure the rate of coincident neutron emission in the measured item. Fig. 1 shows the probability of detecting a neutron as a function of time.

3.2.17 *neutron moderators*, *n*—those materials that slow fast neutrons through elastic scattering. Materials containing hydrogen are the primary example.

3.2.18 *neutron multiplication*, *n*—the fractional increase in the number of second-generation neutrons emitted, following spontaneous fission, due to self-induced fissions in the item being measured.

3.2.19 *passive mode*, *n*—determines the total spontaneously fissioning mass in the measured item through the detection of coincidence neutrons. The coincident neutrons are prompt neutrons.

3.2.20 *predelay*, *n*—the time interval immediately after the detection of the initiating neutron. This time is selected to allow the electronics to recover and detect subsequent neutrons.

3.2.21 *prompt and delayed neutrons*, *n*—neutrons occurring as a result of fissions. Approximately 99 % are prompt neutrons, emitted directly from fission within 10^{-13} s after fission begins. The remainder are delayed neutrons, the result of neutron decay by some of the fission products. Delayed neutrons appear seconds or minutes after the fission begins.

3.2.22 *(α, n) reactions*, *n*—occur when energetic alpha particles collide with low atomic number nuclei, such as O, F, or Mg, producing single neutrons. Neutrons produced in this manner are not correlated in time and are a source of "singles" in passive neutron counting and a source of background in active neutron counting.

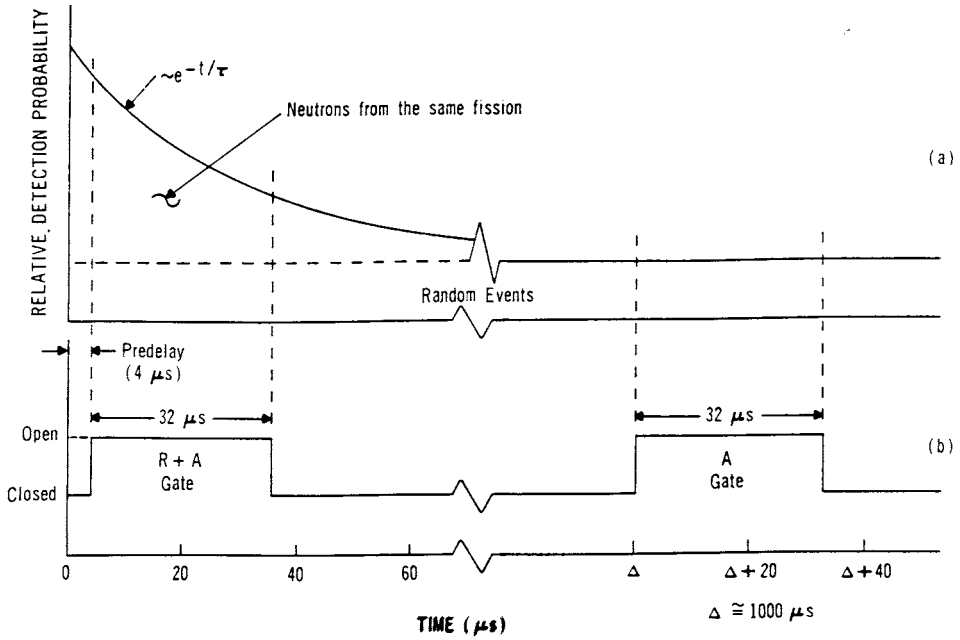
3.2.23 *reals (r)*, *n*—the number of real coincident neutrons in the ($r + a$) gate interval following the initial detection of each neutron during the selected count time, *t*. This quantity is derived from the two measured quantities, $r + a$ and a.

3.2.24 *reals plus accidentals ($r + a$)*, *n*—the number of neutrons detected in the ($r + a$) gate interval following the initial detection of each neutron during the selected count time, *t*. These events are due to neutrons that are coincident with the initial neutron (reals) and to neutrons that are not correlated with the initial neutron (accidental coincidences). This is a measured quantity.

3.2.25 *shift-register-based coincidence circuit*, *n*—an electronic circuit for determining totals ([τ]), reals plus accidentals ($r + a$), and accidentals (a) in a selected count time (t). Fig. 1 illustrates the time relationship between the measured quantities.

3.2.26 *shuffler technique*, *n*—an active-neutron nondestructive assay technique that moves a ^{252}Cf source close to the

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



a) Simplified relative neutron detection probability distribution.
b) Coincidence gate timing diagram.

NOTE 1—Curve (a) is a simplified probability distribution showing the decay, as a function of time, for detecting a second neutron from a fission event. The probability for detecting an uncorrelated neutron is constant with time. Typical coincidence timing parameters are shown in (b).

FIG. 1 Probability of Neutron Detection as a Function of Time

assay item to irradiate the fissile material, then counts delayed neutrons from the induced fissions after the source is withdrawn. Fig. 2 illustrates the measurement concept, and the two source positions that the source "shuffles" between.

3.2.27 *totals ([τ])*, *n*—the total number of individual neutrons detected during the selected count time, *t*. This is a measured quantity.

3.2.28 *transuranic waste (TRU waste)*, *n*— defined by the United States Department of Energy as any waste containing alpha-emitting isotopes with atomic number greater than 92 and half-life greater than 20 years, with α activity concentrations greater than 100 nCi per gram of bulk waste.

3.3 *volume weighted average response, n*—an estimate of the count rate that would be obtained from a drum containing a uniform distribution of special nuclear material. It is a weighted average calculated from a series of measurements as follows:

3.3.1 The drum is divided into 15 or so volume elements,

3.3.2 A point source is centered in one of the volume elements and measured,

3.3.3 The point source is moved to the next volume element and measured, and

3.3.4 Each response is weighted by the size of the corresponding element. (See Appendix X1 for a more detailed explanation.)

4. Summary of Test Method

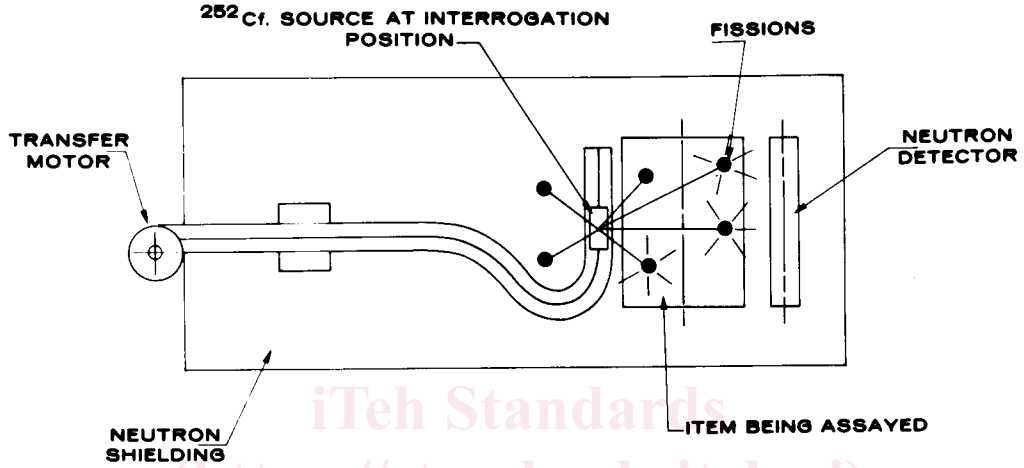
4.1 This test method consists of two distinct modes of operation: passive and active. The instrument that performs the active mode measurement is referred to as a "shuffler" due to the motion of the ^{252}Cf source. This test method usually relies on passive neutron coincidence counting to determine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

4.1.1 *Passive Neutron Coincidence Counting Mode*—The even mass isotopes of plutonium fission spontaneously. Approximately two prompt neutrons are emitted per fission. The number of these coincident neutrons detected by the instrument is correlated to the quantity of even mass isotopes of plutonium. The total plutonium 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 Ref **4** and Test Method C 1207).

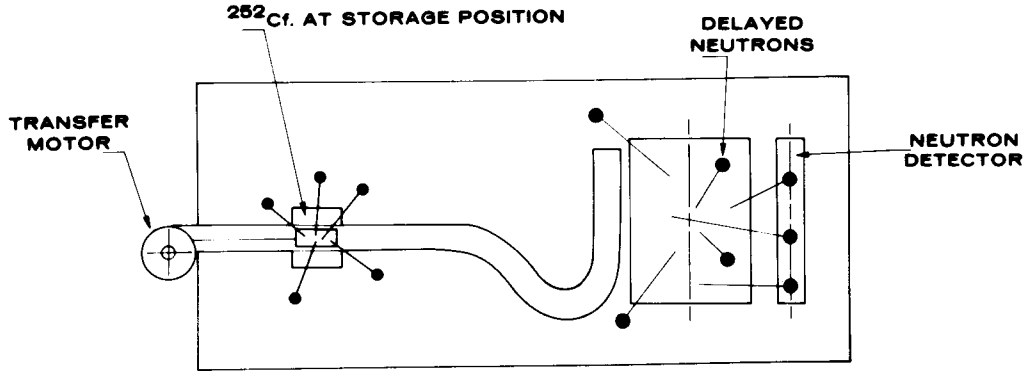
4.1.2 *Active Neutron (Shuffler) Mode*—Fissions in ^{235}U can be induced by bombarding uranium with neutrons. Approximately 1 % of the neutrons per fission are delayed, being emitted from the fission products for several minutes after the fission event. The active mode consists of several irradiate-count cycles, or shuffles, of the ^{252}Cf source between the positions illustrated in Fig. 2. Californium-252 emits a fission neutron spectrum. During each shuffle, a ^{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 these delayed neutrons detected by the instrument is correlated with the quantity of ^{235}U. The total uranium mass is determined from the known isotopic ratios and the measured quantity of ^{235}U.

^{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 two main features of the active technique are shown. The shuffler measurement consists of several cycles. Each cycle includes an irradiation of the item by the ^{242}Cf source for about 10 s, followed by a counting period of about 10 s while the source is stored in a shield.

FIG. 2 ^{252}Cf Shuffler Measurement Principle

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 neutron signal caused by the matrix.

4.3 Corrections are made for electronic deadtime, neutron background, and the ^{252}Cf source decay.

4.4 The active mode also induces fissions in plutonium if it is present in the assay item. The passive measurement of

plutonium can be used to correct the active measurement of ^{235}U for the presence of plutonium.

4.5 Calibrations are based on measurements of well documented reference materials. 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 uranium and plutonium content of scrap and waste in containers. Measurement count times have been 100 to 1000 s. 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-litre drums with plutonium content ranging from 30 mg to 1 kg.

5.1.2 The active measurement has been applied to benign matrices in 208-litre drums with ^{235}U content ranging from 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. (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 can detect diversion attempts that use bulk neutron 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.

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.

5.5 This test method assumes that the 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 large quantities of special nuclear material are not concentrated in a small portion of the container.

5.8 Reliable and consistent results from the application of this test method require training of personnel who package the scrap and waste prior to measurement. (See ANSI 15.20, Guide C 1009, Guide C 986, and Guide C 1068 for training guidance.)

6. Interferences

6.1 Potential sources of measurement interference include unexpected nuclear material contributing to the active or passive neutron signal, self-shielding by large lumps of fissile material, neutron multiplication, excessive quantities of absorbers or moderators in the matrix, heterogeneity of the matrix, and the heterogeneity of the nuclear material distribution 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 **(5)**.

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 responses of those nuclides must be known.

6.2.1 *Active Mode*— The presence of other fissionable nuclides will increase the delayed neutron count rate, causing an overestimation of the ^{235}U content unless a correction is made. For example, a calibration based on highly enriched uranium will cause biased results if the unknowns contain low-enriched uranium or plutonium.

6.2.2 *Passive Mode*— The presence of other spontaneous fission nuclides, such as curium, will increase the coincident neutron rates, causing an overestimation of plutonium content unless a correction is made. The active mode measurement of plutonium is not sensitive to this source of bias.

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

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.

6.3.2 *Passive Mode (Multiplication)*—Neutrons originating in the lump induce fissions in the same lump.

6.4 Moderators 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 **(2)**.

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.

6.4.3 The instrument produces a nonuniform response for large containers with large 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 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 to small quantities of special nuclear material for both the active and passive modes.

7. Apparatus

7.1 The apparatus used in this test method can be obtained commercially. Specific applications may require customized designs. The following description is one possible design. Fig. 3 is a cutaway illustration of a shuffler to measure 208-litre 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.

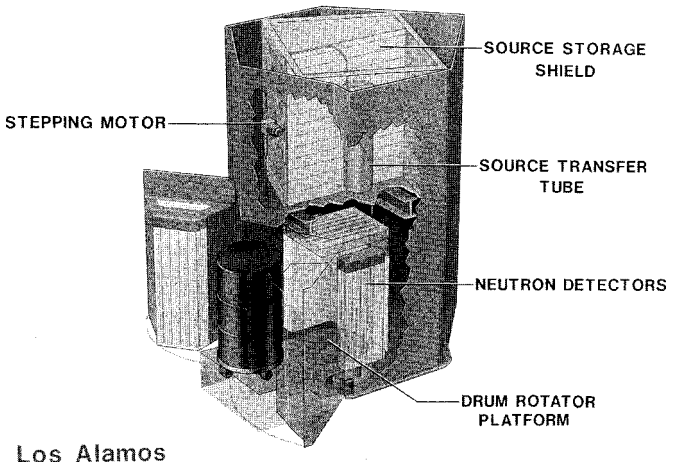
7.2 *Counting Assembly*—see Fig. 4.

7.2.1 The neutron detectors are ^{3}He proportional counters embedded in polyethylene, located around the item in a near 4π geometry. The detection efficiency for neutrons of fission energy should be at least 15 %. Larger detection efficiencies provide better precision and lower detection limits for a given count time. 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 proportional counters mounted on the inner walls of the measurement chamber and not embedded in polyethylene. One flux monitor is covered with cadmium 1 mm thick; the other is bare. The cadmium shields one flux monitor from thermal neutrons; therefore, the two flux monitors can be compared in order to provide information about the neutron energy distribution.

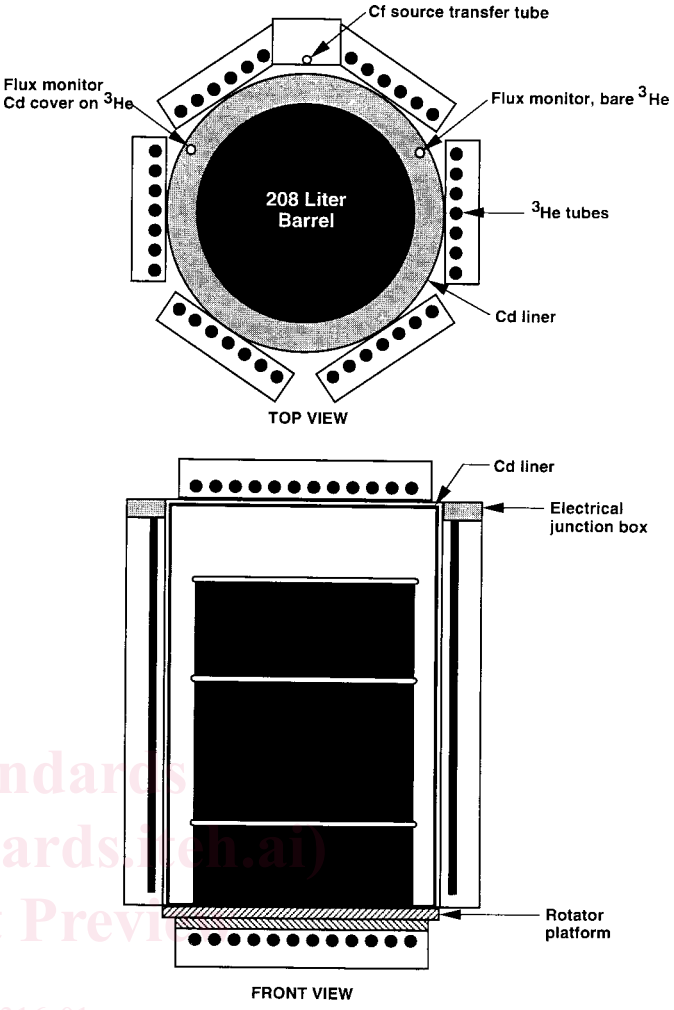
7.3 *Shielding*—The quantity of radiation shielding for the ^{252}Cf source is determined by personnel safety requirements rather than by background considerations.

7.3.1 The measurement chamber is surrounded by one or two feet of materials such as polyethylene and boron to shield the operator during the ^{252}Cf irradiation.



NOTE 1—A sketch of a shuffler designed to assay 208-litre drums. The source storage shield is a 2000-kg, 1.2-metre cube that resides close to the measurement chamber. In this design it is on top of the measurement chamber. The stepping motor pushes the ^{252}Cf source through the source transfer tube between the storage position and the irradiation position inside the measurement chamber.

FIG. 3 Shuffler for 208-litre Drums of Waste



NOTE 1—The front and top views of the measurement chamber shown in Fig. 1 are shown here in detail. The 208-litre barrel rotates on a platform above the bottom detector bank. Six side banks surround the drum, with the ^{252}Cf source transfer tube at the rear of the item. The two flux monitors are placed at the rear of the item chamber.

FIG. 4 Shuffler Detector Bank Diagram

7.3.2 The shield for the ^{252}Cf storage position is typically 0.6 m thick (1.2-m cube), or the source is placed 1.8 m underground. Composite shields are more effective than polyethylene for large ^{252}Cf sources **(6)**.

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

7.5 *Californium-252 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 that offers precise timing, positioning, and computer control. During the active measurement, variations in the