



Designation: C1458 – 16

Standard Test Method for Nondestructive Assay of Plutonium, Tritium and ²⁴¹Am by Calorimetric Assay¹

This standard is issued under the fixed designation C1458; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ϵ) indicates an editorial change since the last revision or reapproval.

1. Scope

1.1 This test method describes the nondestructive assay (NDA) of plutonium, tritium, and ²⁴¹Am using heat flow calorimetry. For plutonium the typical range of applicability, depending on the isotopic composition, corresponds to ~0.1 g to ~5 g quantities while for tritium the typical range extends from ~0.001 g to ~400 g. This test method can be applied to materials in a wide range of container sizes up to 380 L. It has been used routinely to assay items whose thermal power ranges from 0.001 W to 135 W.

1.2 This test method requires knowledge of the relative abundances of the plutonium isotopes and the ²⁴¹Am/Pu mass ratio to determine the total plutonium mass.

1.3 This test method provides a direct measure of tritium content.

1.4 This test method provides a measure of ²⁴¹Am either as a single isotope or mixed with plutonium.

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

1.6 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

2. Referenced Documents

2.1 ASTM Standards:²

C697 Test Methods for Chemical, Mass Spectrometric, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powders and Pellets

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

Current edition approved March 1, 2016. Published April 2016. Originally approved in 2000. Last previous edition approved in 2009 as C1458 – 09^{ε1}. DOI: 10.1520/C1458-16.

² 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.

C1009 Guide for Establishing and Maintaining a Quality Assurance Program for Analytical Laboratories Within the Nuclear Industry

C1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry

C1592 Guide for Nondestructive Assay Measurements

C1673 Terminology of C26.10 Nondestructive Assay Methods

2.2 ANSI Standard:³

ANSI N15.36 Measurement Control Program – Nondestructive Assay Measurement Control and Assurance

3. Terminology

3.1 Definitions:

3.1.1 Terms shall be defined in accordance with C26.10 Terminology C1673 except for the following:

3.1.2 *active mode*—a mode of calorimeter operation where an external power, applied by means of a series of band heaters for instance, is used to maintain the calorimeter at constant temperature.

3.1.3 *baseline, n*—the calorimeter output signal with no heat-generating item in the calorimeter item chamber.

3.1.4 *base power, n*—a constant thermal power applied in a calorimeter through an electrical resistance heater with no heat-generating item in the item chamber.

3.1.5 *equilibrium, n*—the point at which the temperature of the calorimeter measurement cell and the item being measured stabilizes.

3.1.6 *heat distribution error, n*—the bias arising from the location of the heat source within the calorimeter chamber.

3.1.7 *passive mode, n*—a mode of calorimeter operation where temperature change (caused by the heat to be quantified) is measured by means of temperature sensors with no external power applied except in the case of Wheatstone bridge temperature sensors where electrical current is needed to excite the bridge circuit.

³ Available from American National Standards Institute (ANSI), 25 W. 43rd St., 4th Floor, New York, NY 10036, http://www.ansi.org.

3.1.8 *power compensation mode, n*—a mode of calorimeter operation where a constant applied thermal power is maintained in a calorimeter measurement chamber through the use of an electric resistance heater in a closed loop control system. Historically, Mound Laboratory used to call this “Servo Control.”

3.1.9 *sensitivity, n*—the change in calorimeter response per Watt of thermal power (usually in units of $\mu\text{V}/\text{W}$) for a heat flow calorimeter.

3.1.10 *specific power, n*—the rate of energy emission by ionizing radiation per unit mass of a radionuclide, such as ^{241}Am or tritium.

3.1.11 *thermal diffusivity, n*—the ratio of thermal conductivity to the heat capacity. It measures the ability of a material to conduct thermal energy relative to its ability to store thermal energy.

3.1.12 *thermal power, n*—the rate at which heat is generated in a radioactively decaying item.

3.1.13 *thermal resistance, n*—ratio of the temperature difference at two different surfaces to the heat flux through the surfaces at equilibrium.

3.1.14 *thermal time constant, n*—an exponential decay constant describing the rate at which a temperature approaches a constant value. The combination of the item and its container will have numerous thermal time constants.

4. Summary of Test Method

4.1 The item is placed in the calorimeter measurement chamber and the heat flow at equilibrium, that is, the thermal power, from the item is determined by temperature sensors and associated electronic equipment.

4.2 The thermal power emitted by a test item is directly related to the quantity of the radioactive material in it. The power generated by ionizing radiation absorbed in the item is measured by the calorimeter.

4.3 The total power W_{item} from a mixture of isotopes in the test item is the sum of the power from each heat-producing isotope:

$$W_{item} = \sum_i m_i \cdot P_i \quad (1)$$

where m_i is the mass of the i^{th} isotope and P_i is the specific power (W/g isotope) with the sum taken over all heat-producing isotopes, most usually ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu , and ^{241}Am for Pu-bearing items

4.4 The mass of Pu, tritium, or ^{241}Am is calculated from the measured thermal power of an item, refer to 11.3.

4.4.1 When tritium is the only heat source the measured thermal power can be directly converted into the tritium mass using the specific power of tritium, $P_{eff} = (0.3240 \pm 0.00045)$ (SD) W/g (1).⁴

4.4.2 When ^{241}Am is the only heat source, as a single isotope, the measured thermal power can be directly converted into mass using the specific power of ^{241}Am , $P_{eff} = (0.1142 \pm 0.00042)$ (SD) W/g (see Table 1).

4.4.3 The ^{241}Am mass, m_{Am} , in a plutonium-bearing item is determined by multiplying the Pu mass by the Am/Pu mass ratio. The Am/Pu mass ratio is typically determined by gamma-ray spectroscopy.

5. Significance and Use

5.1 This test method is considered to be the most accurate NDA technique for the assay of many physical forms of Pu. Isotopic measurements by gamma-ray spectroscopy or destructive analysis techniques are part of this test method when it is applied to the assay of Pu.

5.1.1 Calorimetry has been applied to a wide variety of Pu-bearing solids including metals, alloys, oxides, fluorides, mixed Pu-U oxides, mixed oxide fuel pins, waste, and scrap, for example, ash, ash heels, salts, crucibles, and graphite scarfings) (2, 3). This test method has been routinely used at U.S. and European facilities for Pu process measurements and nuclear material accountability since the mid 1960's (2-9).

5.1.2 Pu-bearing materials have been measured in calorimeter containers ranging in size from about 0.025 m to about 0.63 m in diameter and from about 0.076 m to about 1.38 m in height.

5.1.3 Gamma-ray spectroscopy typically is used to determine the Pu isotopic composition and ^{241}Am to Pu ratio (see Test Method C1030). However, isotopic information from mass spectrometry and alpha counting measurements may be used instead (see Test Method C697).

5.2 This test method is considered to be the most accurate NDA method for the measurement of tritium. For many physical forms of tritium compounds calorimetry is currently the only practical measurement technique available.

5.3 Physical standards representative of the materials being assayed are not required for the test method.

5.3.1 This test method is largely independent of the elemental distribution of the nuclear materials in the matrix.

5.3.2 The accuracy of the method can be degraded for materials with inhomogeneous isotopic composition.

5.4 The thermal power measurement is traceable to national measurement systems through electrical standards used to directly calibrate the calorimeters or to calibrate secondary ^{238}Pu heat standards.

5.5 Heat-flow calorimetry has been used to prepare secondary standards for neutron and gamma-ray assay systems (7-12).

5.6 Four parameters of the item and the item packaging affect measurement time. These four parameters are density, mass, thermal conductivity, and change in temperature. The measurement well of passive calorimeters will also affect measurement time because it too will need to come to the new equilibrium temperature. Calorimeters operated in power compensation mode maintain a constant measurement well temperature and have no additional effect on measurement time.

5.6.1 Calorimeter measurement times range from 20 minutes (13) for smaller, temperature-conditioned containers up to 72 h (14) for larger containers and items with long thermal-time constants.

5.6.2 Measurement times may be reduced by using equilibrium prediction techniques, by temperature preconditioning of

⁴ The boldface numbers in parentheses refer to the list of references at the end of this standard.

the item to be measured, by operating the calorimeter using the power compensation technique, or by optimization of the item container (low thermal mass and high thermal conductivity) and packaging.

6. Interferences

6.1 For plutonium-bearing items alpha decay heating is usually the dominant heat source and any interferences are typically negligible. These minor effects included the points 6.2 and 6.3.

6.2 Interferences for calorimetry are those processes that would add or subtract thermal power from the power of the radionuclides being assayed. Some examples include phase changes, endothermic or exothermic chemical reactions, such as oxidation, radiolysis of liquids, and bacterial action.

6.3 For the P_{eff} calculation, charged particles are assumed to be totally absorbed in the matrix. The contribution of high energy gamma-rays (for example, 1.173 MeV and 1.333 MeV of the ^{60}Co) needs to be calculated to not underestimate their contribution which could bias the measurement results. These assessments requiring subject matter expertise are usually based on high-resolution gamma spectroscopy (HRGS) and need to be considered in a case by case basis. These effects are usually considered negligible (15, 16).

6.4 The loss of energy by escaping neutrons following spontaneous and induced fissions is about 1%. The loss of energy by escaping neutrons following (α, η) reactions is around 0.007% per alpha particle. These effects are also usually considered negligible (15, 16).

7. Apparatus

7.1 Calorimeters are designed to measure different sizes and quantities of nuclear material. Different types of heat-flow calorimeter systems share the common attributes listed below.

7.1.1 *Measurement Chamber*—Heat flow calorimeters typically have a cylindrical, cuboid, or hexagonal measurement chamber from which all of the heat flow generated by radioactive decay is directed through temperature sensors. It may also have a reference chamber in which a dummy can is placed (so-called twin cell calorimeter) (15, 17, 18).

7.1.1.1 An electrical heater may be built into the walls or the base of the chamber to provide measured amounts of thermal power into the calorimeter well.

7.1.1.2 Insulation or active heaters (or both) should surround the cavity to shield the chamber from outside temperature variations that would influence the thermal power measurement. Typically, an insulated plug or a temperature-controlled plug with an independent electrical heater is inserted above the item container inside the calorimeter. For some calorimeter types an insulating plug is installed permanently below the measurement chamber.

7.1.2 *Calorimeter Can*—The item to be measured may be placed in a special can that is designed to be inserted and removed easily from the calorimeter. It will typically have only a small air gap to provide good thermal conductivity between the outer surface of the can and the inner surface of the measurement chamber.

7.1.3 *Temperature Sensors*—Temperature sensors consist of thermistors, thermocouples, temperature sensitive resistance wires, thermopiles, or Peltier modules working in Seebeck mode. Temperature sensors should be placed as close as possible in a geometrical homogeneous network all around the can.

7.1.4 *Thermal Sink*—The temperature increases due to heat flows generated by items are measured against a reference temperature of a thermal sink. The thermal sink could be a water bath, air bath, a solid block, usually metallic block, maintained at a constant temperature or a combination of a solid block and water bath or air bath.

7.1.5 *Electrical Components*—Sensitive, stable electronic components are required for accurate calorimeter measurements.

7.1.5.1 High precision voltmeters or voltage measurement devices are required to quantify the voltage changes generated from the temperature sensors. The resolution should be better than one part per million of the voltage range.

7.1.5.2 Stable power supplies are necessary to provide constant current to Wheatstone bridge sensors and calorimeter heaters.

7.1.5.3 Precision resistors with certified resistances traceable to a national measurement system may be used with calibrated voltmeters to accurately determine electrical power delivered to heaters in the calorimeter chamber. If radioactive heat standards are used as part of the measurement control program the calorimeter voltmeters need not be calibrated nor are precision resistors required.

7.1.5.4 For a calorimeter operated in the power compensation mode digital-to-analog controller units are used to supply power to an internal resistance heater to maintain a constant temperature differential across thermal resistances.

7.1.6 *Heat Standards*—Thermal power standards are required to calibrate the calorimeter and may be used as measurement control standards to check the stability of calorimeter performance (19-22).

7.1.6.1 Radioactive heat standards, typically ^{238}Pu heat sources, also may be used to calibrate calorimeters over a range of thermal powers. These standards are calibrated against electrical standards traceable to a national measurement system. The certified power is typically decay corrected to the nearest day using certified decay tables.

7.1.6.2 Removable electrical heaters may be used to calibrate calorimeters. For this type of standard the power generated by the heater must be measured with electrical equipment regularly calibrated against standards or standard methods traceable to a national measurement system. The power supplied to the electrical calibration heater may be varied over the calibration range.

7.1.7 *Wheatstone Bridge*—When temperature sensitive resistance wire is used as the sensor, it is arranged in a Wheatstone bridge configuration shown in Fig. 1.

7.1.8 *Data Acquisition System*—Calorimeter data collection is performed using computer-based data acquisition systems. The system should be able to read signal voltages or resistances at a fixed time frequency and be able to calculate and report a

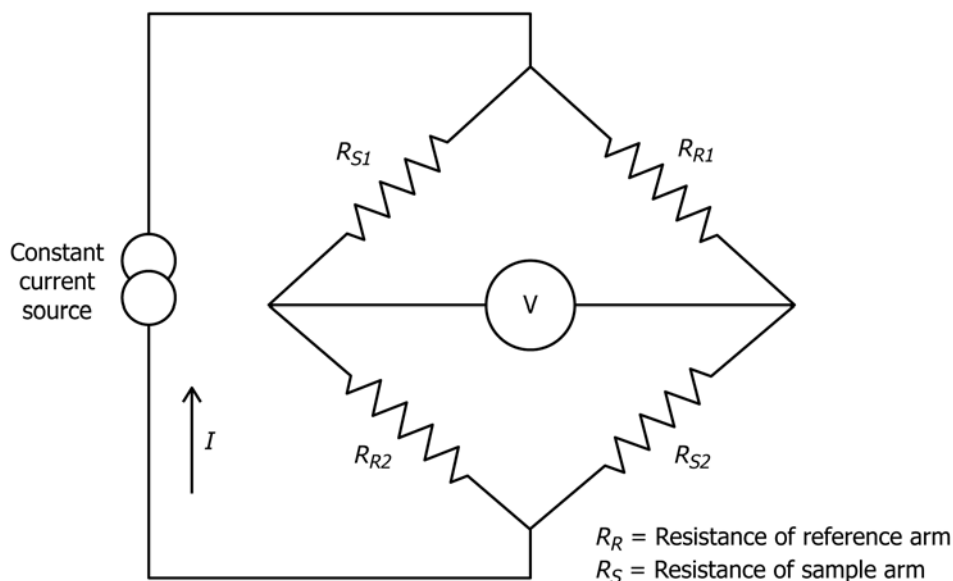


FIG. 1 Calorimeter Wheatstone Bridge Circuit

power value from the item using software that detects equilibrium. Graphics and numerical data indicating system power and temperatures may be displayed to aid the operator.

7.1.9 *Adapters*—Low mass cylindrical metal adapters may be fabricated to accept smaller calorimeter containers in the calorimeter well, and thus, provide good thermal contact between the outer container surface and calorimeter inner wall. Heat-conducting metal foil or metal gauze fill material, typically Al or Cu, or metal shot can be used in place of machined metal adapters. Smaller items may be placed in the calorimeter container and the void space inside the container may be filled with metal fill material or shot to provide good thermal contact. Lack of fill material does not preclude measurement of the item but it will increase the time to reach the calorimetric equilibrium and thus increase the total measurement time.

7.1.10 *Loading Apparatus*—A hoist or other assist may be used to load and unload items. Robotic loading systems may be used to handle the items.

8. Heat-Flow Calorimeter Systems

8.1 A heat flow calorimeter system consists of an item chamber thermally insulated from a constant temperature environment by a thermal resistance. It may also include a reference chamber (twin calorimeter) also insulated from a constant temperature environment by a thermal resistance (15, 17, 18). When an item is placed in the calorimeter the temperature difference across the thermal resistance is disturbed and the difference changes with time until it converges to a constant value and equilibrium is achieved. The magnitude of the shift in the measured voltage (passive mode) or supplied power (power compensation mode) is used to determine the thermal power of the item in the calorimeter.

8.1.1 The curve of the temperature difference displaying the approach to the calorimetric equilibrium is a function of several exponentials with different time constants related to the specific heats and thermal conductivities of the item matrix material, packaging, and the calorimeter.

8.1.1.1 Equilibrium may be detected by visual inspection of the measurement data versus time or through statistical tests performed on a set of the latest data points in the time series.

8.1.1.2 Statistical prediction algorithms may be used during transient temperature conditions to predict equilibrium and reduce measurement time. These typically consist of exponential functions that are used to fit the measurement data. The fitted parameters then are used to predict the final equilibrium power.

8.1.1.3 The temperature of the item to be measured may be adjusted through the use of preconditioning heaters or baths in order to decrease the time required to reach equilibrium.

8.2 Heat-flow calorimeters are operated typically in one of two modes, passive mode or power compensation mode.

8.2.1 *Passive Mode Operation*—A plot with an example of a passive mode calorimeter response to a heat source is shown in Fig. 2. This plot shows that after a period of time the temperature transient caused by the insertion of the item into the calorimeter disappears and the calorimeter and item are in thermal equilibrium after approximately 7 h in this example.

8.2.1.1 The item power, W_{item} , is calculated by the following:

$$W_{item} = (BP_s - BP_0)/S \quad (2)$$

where:

- S = the calorimeter sensitivity ($\mu V/W$) at the power level, determined by electrical or ^{238}Pu standards,
- BP_s = the equilibrium sensor response with the item in the calorimeter, and
- BP_0 = the baseline sensor response with no item in the calorimeter.

8.2.2 *Power Compensation Mode*—In this mode of operation, a constant amount of thermal power is applied to the item chamber by electrical heaters. The temperature of the calorimeter item chamber is held at a constant temperature difference above the temperature of the thermal sink by means

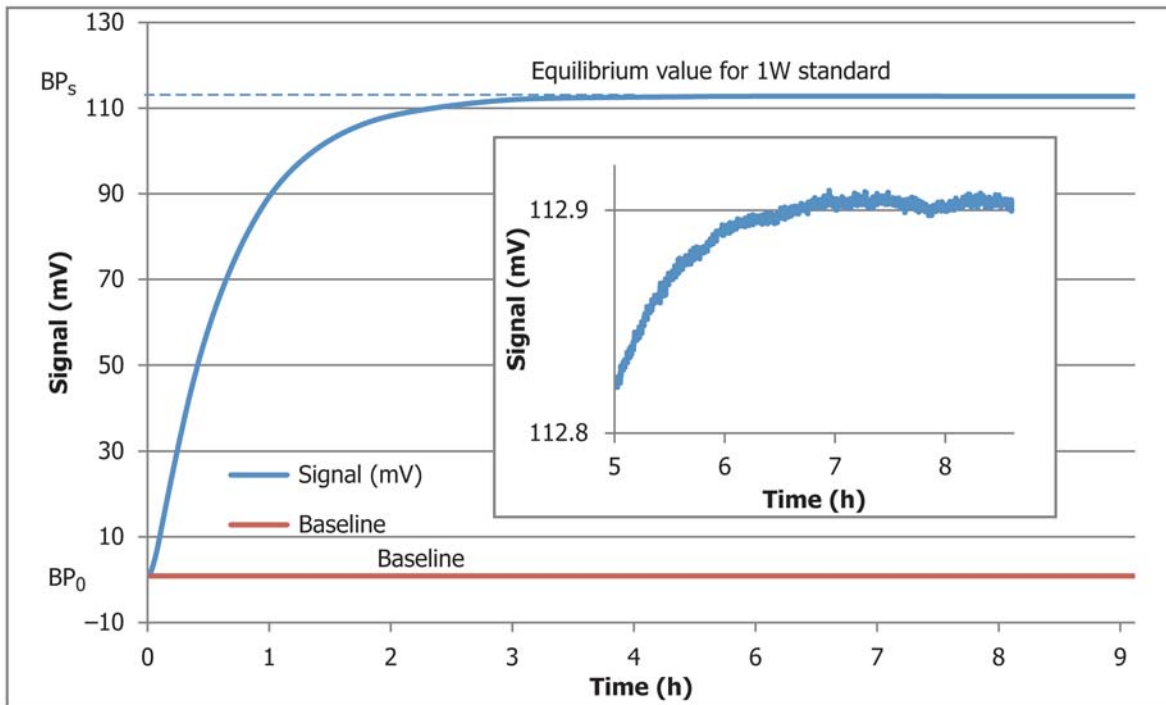


FIG. 2 Approach to Equilibrium for a Calorimeter in the Passive Mode

of a controlled power unit. The constant power causes a constant temperature differential to be maintained across the thermal resistance separating the measurement chamber from a controlled reference temperature. The temperature differential is proportional to the signal, voltage or resistance, and is the temperature difference between a sensor (or sensors) located adjacent to the item being measured and the other(s) located at the reference temperature. A closed-loop controller monitors the output signal, and if a radioactive heat-generating item is

inserted, the external power applied is decreased to precisely maintain the same signal differential. When the unknown item is placed in the calorimeter, the control power drops over time to a lower level. The power of the unknown is the difference between the two control power readings at equilibrium. A plot with an example of the calorimeter response is shown in Fig. 3.

8.2.2.1 The item power, W_{item} , is calculated by the following:

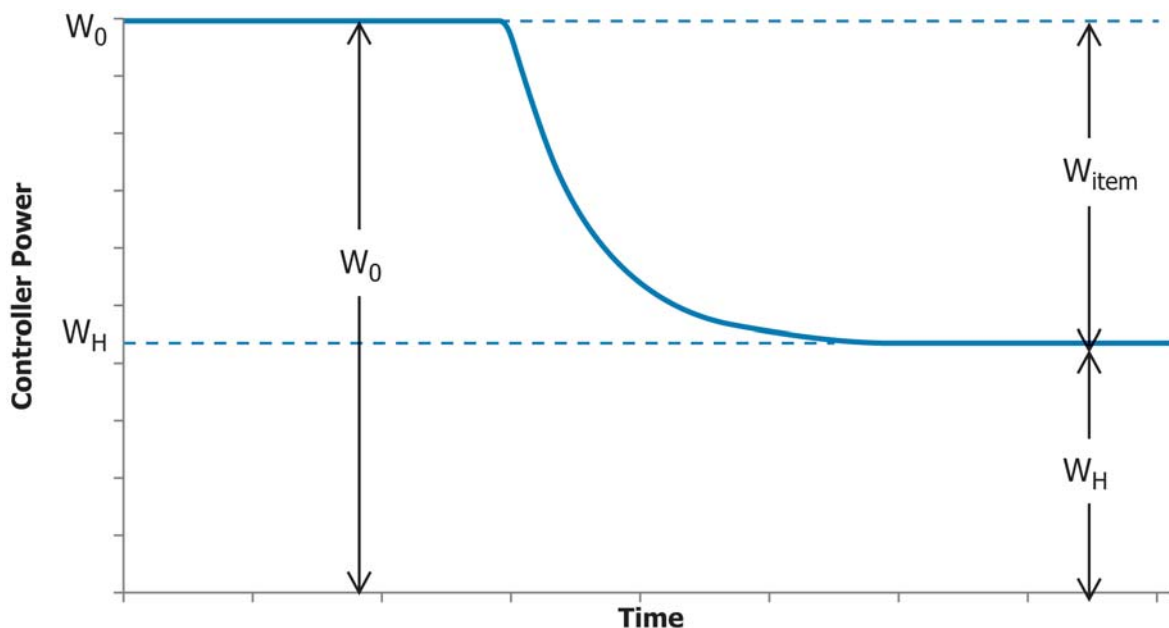


FIG. 3 Approach to Equilibrium for a Calorimeter Operated in the Power Compensation Mode

$$W_{item} = W_0 - W_H \quad (3)$$

where:

W_0 = the base power with no item in the calorimeter, and
 W_H = the power supplied to the calorimeter with the item in the calorimeter.

8.2.2.2 The measurement time for the power compensation mode of operation is normally shorter than for the passive mode because the calorimeter components are at the equilibrium temperature and the controlled internal heater can supply heat actively to bring the item to equilibrium.

9. Hazards

9.1 Safety Hazards:

9.1.1 It is recommended that a criticality evaluation be carried out if fissile material is to be measured.

9.1.2 Precautions should be taken to minimize electrical shock hazards.

9.1.3 Precautions should be taken to avoid contamination with radioactive materials.

9.1.4 Precautions should be taken to minimize personnel radiation exposure to ionizing radiation.

9.1.5 Pinch-point and lifting hazards may be present during the loading and unloading of heavy items with calorimeters. Mechanical aids, such as a hoist or any other assist, should be used for movement of heavy items.

9.1.6 High-power items could present a burn hazard to the operator or damages to the instrument.

9.2 Technical Hazards:

9.2.1 Room temperature variations may affect the stability of the reference temperature and increase the measurement uncertainty.

9.2.2 Using a measurement result outside of the range of the calibration is not recommended.

9.2.3 Care should be taken in the insertion or removal of the calorimeter can so that it is not jammed in the calorimeter well.

9.2.4 Noise in the electronics AC supply power generated by nearby machinery may increase the measurement uncertainty.

9.2.5 The base power for power compensation mode calorimeters must not be less than the highest power expected from items.

9.2.6 Mechanical stress on the item chamber from the weight of the item may cause a bias in the final result.

9.2.7 The calorimeter may exhibit a small heat distribution error dependent on calorimeter design and item characteristics.

9.2.8 The following conditions could extend measurement time.

9.2.8.1 Large masses of material.

9.2.8.2 Items that make poor thermal contact with their containers.

9.2.8.3 Items that contain a large amount of insulating material or dead air spaces caused by several layers of containment.

9.2.9 Deviation between the actual approach to equilibrium and end-point prediction algorithm can result in measurement bias.

9.2.10 Bias in the determination of P_{eff} will lead to a bias in the assay result.

9.2.11 For removable electrical heat standards the heater leads to the standard will serve as a heat path for heat to exchange between the measurement chamber and the environment; this may bias the calibration.

9.2.12 Improperly closing the calorimeter (for example, not completely inserting the insulating baffle) may bias the measurement results, degrade precision, or both.

9.2.13 Cables or any other conductor providing a thermal conduction path into and out of the calorimeter may bias the measurement results. Variability in these paths is a more severe hazard.

10. Calibration Procedure

10.1 The type of calibration procedure depends on whether the calorimeter is operated in the passive or power compensation mode. In the passive mode calibration consists of determining the calorimeter sensitivity, S , the conversion factor between the differential voltage or resistance output of the sensor system and the thermal power of the item being measured. In the power compensation mode calibration is setting the sensor output set point voltage that corresponds to a specific base power.

10.2 Calibration—Passive Mode:

10.2.1 Select a series of ^{238}Pu heat standards or calibrated electrical standard power settings that span the expected power range of items to be measured. A minimum of three different standard powers must be used.

10.2.2 Initiate a baseline measurement of the voltage with a calorimeter can, if used, filled with conductive material in the calorimeter chamber. There should be no heat source in the calorimeter can. Record the baseline, BP_0^{pre} , after equilibrium is reached.

10.2.3 Remove the calorimeter can and place the appropriate standard in the measurement cavity as it is done for an unknown item.

10.2.3.1 Whether using radioactive or electrical heat standards, the calorimeter can must be removed from the calorimeter between each measurement, baseline, or standard. If possible, this removal is necessary even when using electrical standards to simulate as closely as possible real calorimeter operating conditions.

10.2.4 Close the calorimeter can and place it in the calorimeter well. For all measurements, the calorimeter should be properly closed before a measurement is commenced.

10.2.5 Initiate the calorimeter run. Record the voltage output (BP_{std}) after equilibrium has been achieved.

10.2.6 Remove the calorimeter can from the calorimeter, then remove the heat standard from the can and place the can back in the calorimeter.

10.2.7 Re-measure the baseline BP_0^{post} after equilibrium is reached as needed.

10.2.8 Use the known power output of the heat standard, W_{std} , the calorimeter sensor value, BP_{std} , and the calculated average baseline (average of BP_0^{pre} and BP_0^{post}), BP_0 to calculate the calorimeter sensitivity, S .