



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

This standard is issued under the fixed designation C 1458; 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 ^{241}Am using heat flow calorimetry. For plutonium the range of applicability corresponds to < 1 g to > 2000 g quantities while for tritium the range extends from 0.001 g to > 10 g. This test method can be applied to materials in a wide range of container sizes up to 50 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 $^{241}\text{Am}/\text{Pu}$ 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 ^{241}Am either as a single isotope or mixed with plutonium.

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.*

2. Referenced Documents

2.1 ASTM Standards:

C 697 Test Methods for Chemical, Mass Spectrometry, and Spectrochemical Analysis of Nuclear-Grade Plutonium Dioxide Powder and Pellets²

C 859 Terminology Relating to Nuclear Materials²

C 1009 Guide for Establishing a Quality Assurance Program for Analytical Chemistry Laboratories Within the Nuclear Industry²

C 1030 Test Method for Determination of Plutonium Isotopic Composition by Gamma-Ray Spectrometry²

2.2 ANSI Standards:³

ANSI N15.22 Plutonium—Bearing Solids—Calibration

Techniques for Calorimetric Assay

ANSI N15.54 Radiometric Calorimeters—Measurement Control Program

3. Terminology

3.1 *Definitions*—Terms shall be defined in accordance with Terminology C 859 except for the following:

3.1.1 *baseline*—the calorimeter output signal with no heat-generating item in the calorimeter sample chamber.

3.1.2 *basepower*—a constant thermal power applied in a calorimeter through an electrical resistance heater with no heat-generating item in the sample chamber.

3.1.3 *calorimeter*—a device to measure heat or rate-of-heat generation.

3.1.4 *calorimetric assay*—determination of the mass of radioactive material through the measurement of its thermal power by calorimetry and the use of nuclear decay constants and, if necessary, additional isotopic measurements.

3.1.5 *effective specific power*—the rate of energy emission per unit mass of plutonium at the time of measurement.

3.1.6 *equilibrium*—the point at which the temperature of the calorimeter measurement cell and the item being measured stops changing.

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

3.1.8 *heat-flow calorimeter*—a calorimeter so constructed that the heat generated in the calorimeter flows past a temperature sensing element, through a thermal resistance, to a constant temperature heat sink.

3.1.9 *passive mode*—a mode of calorimeter operation where no external power is applied to the calorimeter except the current needed to excite the Wheatstone Bridge circuit.

3.1.10 *sensitivity*—the change in calorimeter response per Watt of thermal power (usually in units of micro Volts per Watt) for a heat flow calorimeter.

3.1.11 *servo control*—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.

3.1.12 *specific power*—the rate of energy emission by ionizing radiation per unit mass of an isotope, such as ^{241}Am or tritium.

¹ This test method is under the jurisdiction of ASTM Committee C-26 on Nuclear Fuel Cycle and is the direct responsibility of Subcommittee C26.10 on Nondestructive Analysis.

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² *Annual Book of ASTM Standards*, Vol 12.01.

³ Available from American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

3.1.13 *thermal diffusivity*—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.

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

3.1.15 *thermal time constant*—an exponential decay constant describing the rate at which a temperature approaches a constant value.

3.1.16 *thermel*—the THERMal ELeMent of the calorimeter, including the sample chamber, and temperature sensor.

3.1.17 *traceability*—relating individual measurements through an unbroken chain of calibrations to U.S. or international primary reference materials or to accepted values of fundamental physical constants.

4. Summary of Test Method

4.1 The item is placed in the calorimeter measurement chamber and the total 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 radioactive material in it. The total power generated by ionizing radiation absorbed in the item is captured by the calorimeter.

4.3 The mass of plutonium, tritium, or ^{241}Am (m) is calculated from the measured thermal power of an item (W_i) using the following relationship:

$$m = \frac{W_i}{P_{\text{eff}}} \quad (1)$$

where:

P_{eff} = the effective specific power calculated from the isotopic composition of the item (see Appendix X1 for details of the calculation of P_{eff} for plutonium).

4.3.1 For tritium the measured thermal power can be directly transformed into mass using the specific power of tritium, $P_{\text{eff}} = 0.3240 \pm 0.00045$ (SD) W/g (1).⁴

4.3.2 For ^{241}Am as a single isotope the measured thermal power can be directly transformed into mass using the specific power of ^{241}Am , $P_{\text{eff}} = 0.1142 \pm 0.00042$ (SD) W/g (see Table X1.1).

4.3.3 For ^{241}Am mixed with plutonium, the ^{241}Am mass, M_{Am} , is determined by

$$M_{\text{Am}} = R_{\text{Am}} M_{\text{Pu}} \quad (2)$$

where:

R_{Am} = the mass ratio of ^{241}Am to Pu, and

M_{Pu} = the mass of plutonium.

5. Significance and Use

5.1 This test method is presently the most accurate NDA technique for the assay of many physical forms of plutonium. Isotopic measurements by gamma-ray spectroscopy or destruc-

tive analysis techniques are part of the test method when it is applied to the assay of plutonium.

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). The test method has been routinely used at U.S. and European facilities for plutonium process measurements and nuclear material accountability for the last 30 years (2-6).

5.1.2 Plutonium-bearing materials have been measured in calorimeter containers ranging in size from 0.025 to 0.30 m in diameter and from 0.076 to 0.43 m in height.

5.1.3 Gamma-ray spectroscopy typically is used to determine the plutonium isotopic composition and $^{241}\text{Am}/\text{Pu}$ ratio (see Test Method C 1030). Isotopic information from mass spectrometry and alpha counting measurements may be used (see Test Method C 697).

5.2 The test method is the most accurate NDA method for the measurement of tritium. For many physical forms of tritium compounds calorimetry is the only practical measurement technique available.

5.3 Unlike other NDA techniques no physical standards representative of the materials being assayed are required for the test method.

5.3.1 The 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 the U.S. or other 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).

5.6 The calorimetry measurement times are typically longer than other NDA techniques. The thermal diffusivity of the matrix of the item and its packaging will determine the thermal time constant for heat transfer from the item and hence the measurement time.

5.6.1 Calorimeter measurement times range from 20 minutes (8) for smaller, temperature-conditioned, containers up to 24 h 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 the item to be measured, or operating the calorimeter using the servo-control technique.

6. Interferences

6.1 Interferences for calorimetry are those processes that would add or subtract thermal power from the power of the radionuclides being assayed.

6.2 Interferences can be phase changes or endothermic or exothermic chemical reactions, such as oxidation.

6.3 Undetected heat-generating radionuclides would add additional thermal power to the measurement.

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

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 have a cylindrical measurement chamber from which all of the heat flow generated by radioactive decay is directed through temperature sensors.

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 is used to shield the chamber from outside temperature variations that would influence the thermal power measurement. Typically, an insulated plug 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 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 commercially available thermistors, thermocouples, temperature sensitive resistance wire, or thermopiles.

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 or air bath or a metal block maintained by a thermoelectric cooler/heater. In the case of servo-controlled calorimeters, the measurement chamber is maintained at an elevated temperature compared to the reference temperature.

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

7.1.5.1 High precision voltmeters are required to measure the voltage changes generated from the temperature sensors. The resolution of the voltmeters 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 resistance 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 servo (power replacement) 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 (9-12).

7.1.6.1 Radioactive heat standards, typically powered

by ^{238}Pu , also may be used to calibrate calorimeters over a range of thermal powers. These standards are calibrated against electrical standards traceable to the national measurement system.

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 range of calibration.

7.1.7 *Wheatstone Bridge*—When temperature sensitive resistance wire is used as the sensor, it usually 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 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*—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.

7.1.10 *Loading Apparatus*—A hoist or 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 *Equilibrium*—A heat flow calorimeter consists of a sample chamber thermally insulated from a constant temperature environment by a thermal resistance. 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 (servo mode) is used to determine the thermal power of the item in the calorimeter.

8.1.1 The curve describing the approach to equilibrium of the temperature difference 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, in some instances, 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 earlier 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.

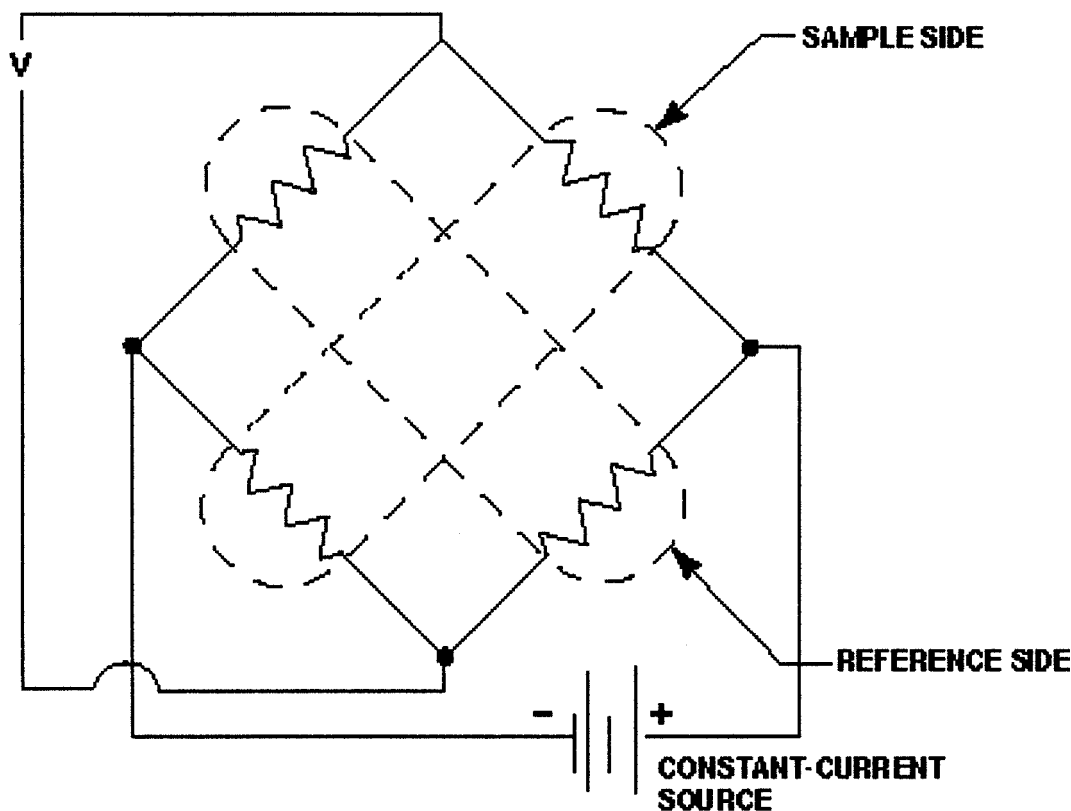


FIG. 1 Calorimeter Wheatstone Bridge Circuit

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

8.2 Heat-flow calorimeters are operated typically in one of two modes, passive or servo controlled (power-replacement).

8.2.1 *Passive Mode*—In this mode of calorimeter operation a Wheatstone Bridge is used as the sensor circuit. The only heat generated comes from the item being measured and the current required to excite the Bridge inside the thermel. A graph of the 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.

8.2.1.1 The item wattage, W_i , is calculated by the following:

$$W_i = (BP_s - BP_0)/S \quad (3)$$

where:

S = the calorimeter sensitivity (microvolts/Watt) determined by electrical or ^{238}Pu standards,

BP_s = the equilibrium bridge potential with the item in the calorimeter, and

BP_0 = the baseline bridge potential with no item in the calorimeter.

8.2.2 *Servo Mode (Power Replacement Mode)*—In this mode of operation, a constant amount of thermal power is applied to the sample chamber by electrical heaters. The temperature of the calorimeter chamber is held several degrees above the temperature of the environment by means of a servo-controller. 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 graph of the calorimeter response is shown in Fig. 3.

8.2.2.1 The item wattage, W_i , is calculated by the following:

$$W_i = W_0 - W_H \quad (4)$$

where:

W_0 = the basepower 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 servo mode of operation is shorter than for the passive mode because the calorimeter components are at the equilibrium temperature and the servo-controlled internal heater can supply heat actively to bring the item to equilibrium.

8.3 *Calorimeter Systems*—A variety of heat-flow calorimeter designs have been used to measure nuclear material. Three types of designs that have been used for accountability measurements are further described below. These are differentiated by the temperature control techniques and heat flow

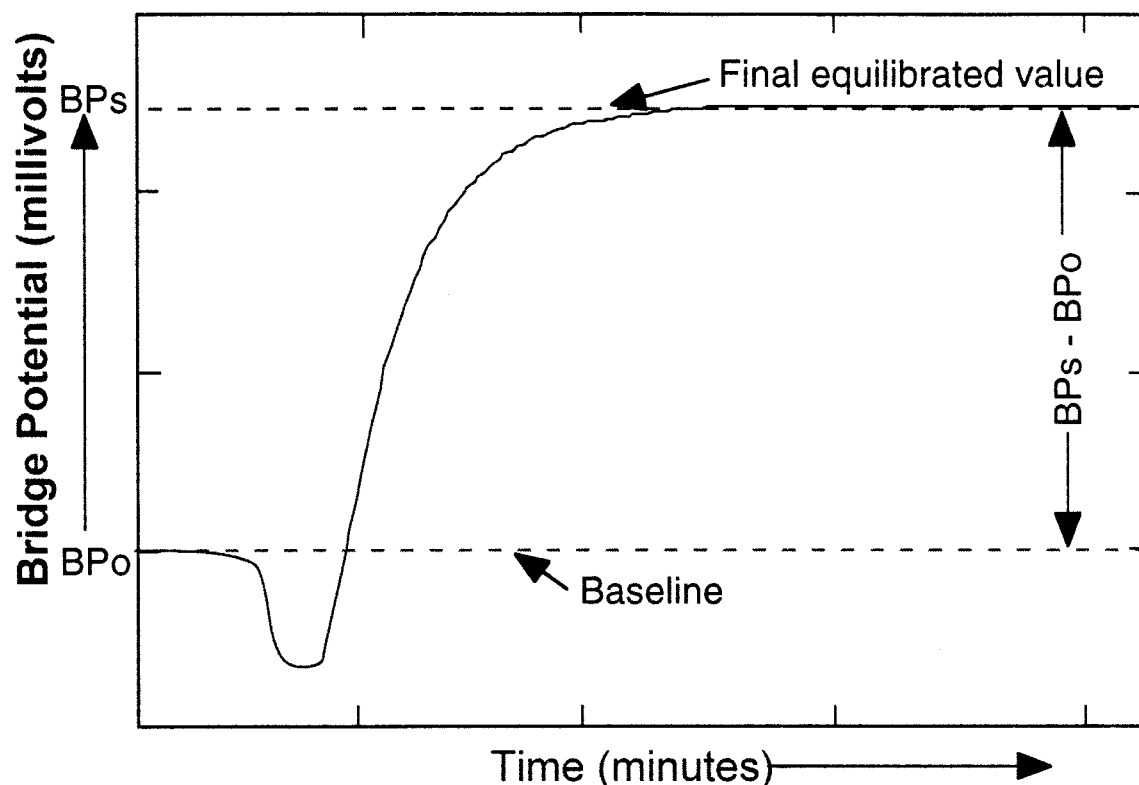


FIG. 2 Approach to Equilibrium for a Calorimeter Operated in the Passive Mode

paths. They are an isothermal “air bath” calorimeter, water bath calorimeter, and rod calorimeter. The air bath and rod calorimeters described below have been operated exclusively in the servo mode, and the water bath calorimeters have been operated in the passive or servo mode.

8.3.1 Isothermal “Air Bath” Calorimeter (12):

8.3.1.1 A schematic diagram of an isothermal (constant temperature) air bath calorimeter is shown in Fig. 4. The calorimeter consists of three concentric cylinders separated by a heat-transfer medium. Each of the cylinders is equipped with temperature sensors. In general, both nickel sense wire and chains of thermistors are used. The outer cylinder is surrounded by a controlled temperature air bath.

8.3.1.2 The temperature sensors are measured using conventional Wheatstone Bridge circuitry or by direct resistance measurement using a high-resolution multimeter. Power to control the temperature of each of the cylinders is supplied by power amplifiers. Heater coils are wound around each cylinder for this purpose.

8.3.1.3 The isothermal calorimeter operates such that each of the three concentric cylinders is at a successively lower temperature as one moves from the inner cylinder (measurement chamber) to the outer cylinder. This difference in temperature results in a temperature gradient and heat flow from the inner cylinder to the outer cylinder.

8.3.1.4 The calorimeter operates in the power replacement mode described in section 8.2.2. The system controller works to maintain the thermal power generated in the measurement chamber at a constant level.

8.3.1.5 The inner cylinder or measurement chamber of the calorimeter is operated at a fixed power known as the base-

power. This basepower must be confirmed periodically as the measurement is based on the difference between the basepower and the final equilibrium or predicted power of the measurement chamber.

8.3.1.6 Isolation from the thermal environment is achieved by circulating air through an exterior chamber either by forced air cooling using room temperature air or by a closed loop air circuit employing a thermoelectric cooling unit to provide a sufficiently cold heat sink temperature.

8.3.1.7 Isothermal calorimeter measurement times may be reduced by item preheating. In this method items are placed in a temperature-controlled chamber that raises their temperature to the operating temperature of the inner cylinder of the calorimeter.

8.3.2 *Water Bath Calorimeter (3)*—Schematics of two different types of water bath calorimeters, the gradient bridge and twin bridge, are shown in Figs. 5 and 6. For both types of calorimeters the Wheatstone Bridge circuit shown in Fig. 1 is used to measure heat flow. The temperature rise in the sample side due to the presence of radioactive material causes the resistance of the sample arms of the Wheatstone Bridge to increase while the resistances of the reference arms remain constant. This causes an imbalance in the bridge and the voltage across the bridge, the bridge potential, changes in proportion to the size of the temperature change. The reference wire arms and sample sensor wire arms of the Wheatstone Bridge are each helically wound interleaved and concentrically around a cylindrical chamber. The windings can be configured in two ways for the gradient-bridge and twin-bridge designs.

8.3.2.1 *Gradient-Bridge Design*—The two-sample sensor wire arms of the Wheatstone Bridge are wound around the

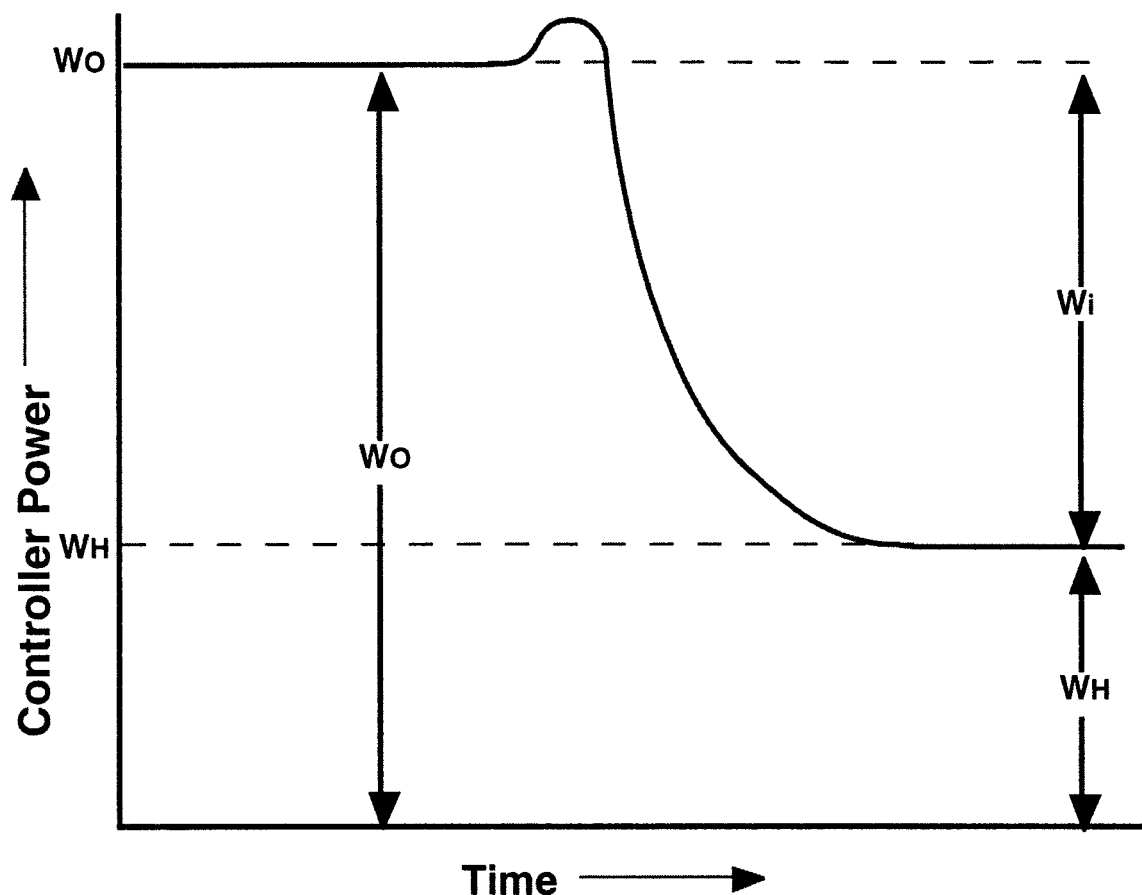


FIG. 3 Approach to Equilibrium for a Calorimeter Operated in the Servo (Power Replacement) Mode

measurement chamber. The reference wire arms are wound concentrically with the sample sensor coils. The two sets of coils are separated by a thermal resistance provided by an air gap or other material. The temperature gradient developed across the thermal gap is proportional to the heat flow from the item. The temperature of the two measurement chamber coils is elevated compared to the reference coils. The two reference coils are maintained at a controlled temperature by a water bath. This type of design normally is operated in the servo mode.

8.3.2.2 Twin-Bridge Design—The two sample arms are wound around the measurement chamber. The two reference wire arms are wound around another chamber identical to the measurement chamber. Both sample and reference sensors are separated by an identical thermal resistance such as an air gap, from a surface maintained at a controlled temperature. The surfaces of cylindrical stainless-steel submarine jackets into which the sample and reference thermels are inserted are maintained at a controlled temperature by a water bath. Twin-bridge calorimeters have been built with the reference thermel located under and coaxial to the sample thermel (“over-under” design) to save space. This type of calorimeter can be operated in the passive or servo mode.

8.3.2.3 The inside of the thermel between the chamber volume and the sensor wire may have a set of heater wires installed. The wires are wound in a grooved metal cylinder called a heater form and are used to introduce known amounts of resistance heat into the chamber. The heater may be used to

operate the calorimeter in the servo mode.

8.3.2.4 A water bath with stirrer or circulating pump is used to maintain a constant reference temperature and serves as a heat sink held at a constant temperature. Water heat sinks can consist of water circulated through an outer jacket (Fig. 5) or a large volume of water in which the calorimeter body is placed (Fig. 6). The twin-bridge design calorimeters are associated with large water baths. Multiple calorimeters have been fit in one water bath. Bath sizes have ranged from 38 to 760 L. The gradient design calorimeters usually have circulating water jackets connected to a separate temperature conditioning system via connecting hoses. Smaller volumes of water, less than 76 L, are required for this type of water bath system.

8.3.2.5 The water bath reference temperature is maintained using temperature controller units. These units may use refrigeration compressors, resistance heaters, thermoelectric cooling units, or evaporative cooling for temperature control. The reference temperature should be controlled to 0.001°C or better.

8.3.2.6 A high-precision voltmeter is used to measure the voltage. This voltmeter is interfaced with a data acquisition computer.

8.3.3 Rod Calorimeter (13):

8.3.3.1 A schematic diagram of the rod calorimeter is shown in Fig. 7. The thermal unit of the calorimeter is comprised of a sample chamber, item insertion/removal plug, thermal shielding, and a highly controlled heat removal path. The heat removal path is through a highly conductive (typically copper)

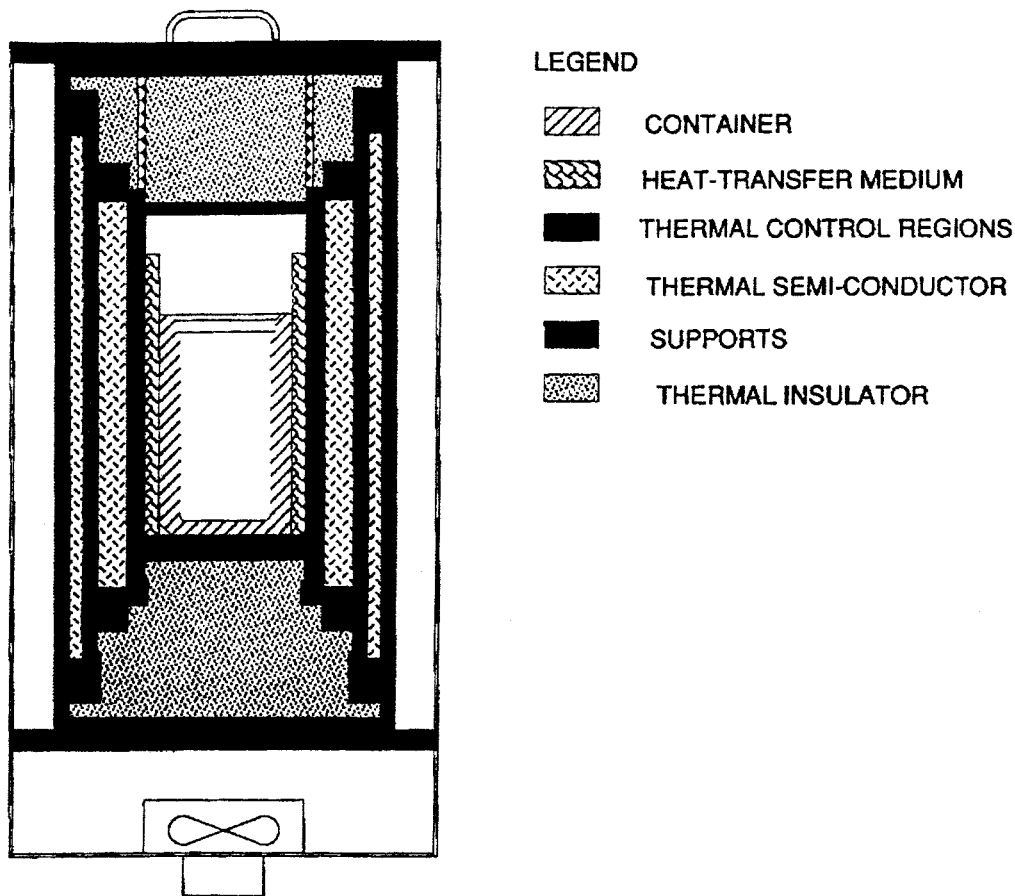


FIG. 4 Isothermal "air bath" Calorimeter

solid rod across which a constant temperature differential is maintained. The upper end of the rod, located at the base of the measurement chamber, is controlled to a constant temperature by supplying heat to the base (or side) of the measurement chamber. The lower end is controlled to a lower temperature, which creates a constant temperature differential. The heat supplied to the base of the measurement chamber is measured. The calorimeter is operated in the servo mode.

8.3.3.2 The thermal shield is composed of several components. The purpose of the thermal shield is to create a zero heat-transfer envelope around the measurement chamber with the exception of a highly controlled heat removal path. Additional constant-temperature thermal shields may be used. Depending on the ambient temperature variations, one or two shields may be incorporated. For lower power measurements, the outermost constant temperature shield typically is a controlled temperature enclosure.

8.3.3.3 Temperature measurements are accomplished using resistance measurements of a thermistor for sensing and a high precision ohmmeter.

8.3.3.4 The plug used to insulate the item being measured is a component of the zero heat-transfer envelope and mitigates thermal effects resulting from gaseous pressure differentials in the measurement chamber.

8.3.3.5 The thermal unit ranges from 4 to 12 closed-loop control systems. Control requires temperature measurement, computer control algorithms with digital-to-analog (D/A) con-

version, and power supplies for driving the heaters and coolers of the thermal unit. The computer D/A outputs are connected to power supplies for driving the heaters/coolers. The power supplies are high grade, low noise, and configured in an operational amplifier mode. System stability analysis is automated and based on power variations and temperature indicators.

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, should be used for movement of heavy items.

9.1.6 A burn hazard can exist for high-power ^{238}Pu items. Caution should be taken to avoid burns.

9.2 Technical Hazards:

9.2.1 Room temperature variations may affect the stability

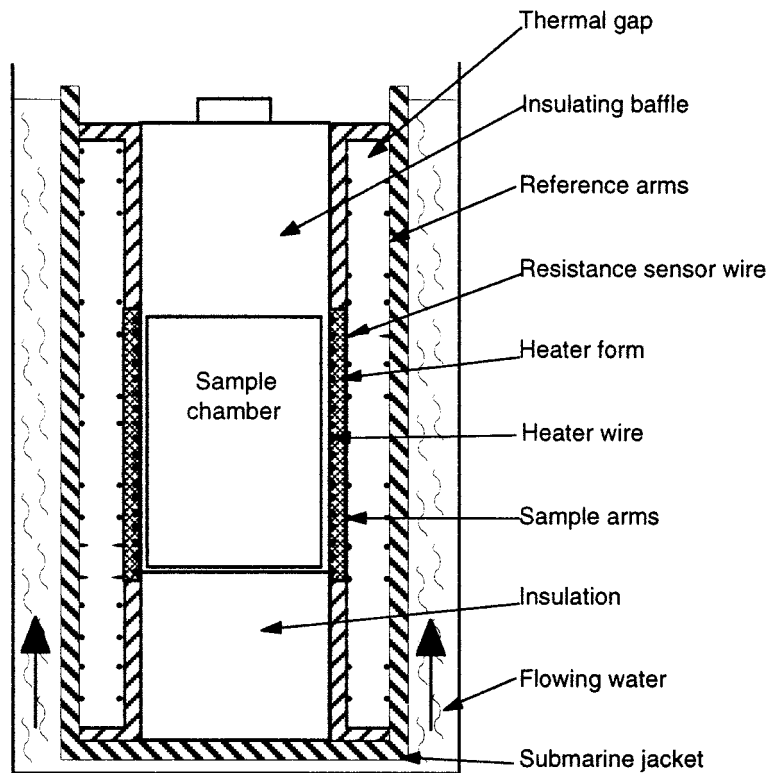


FIG. 5 Gradient Bridge Calorimeter

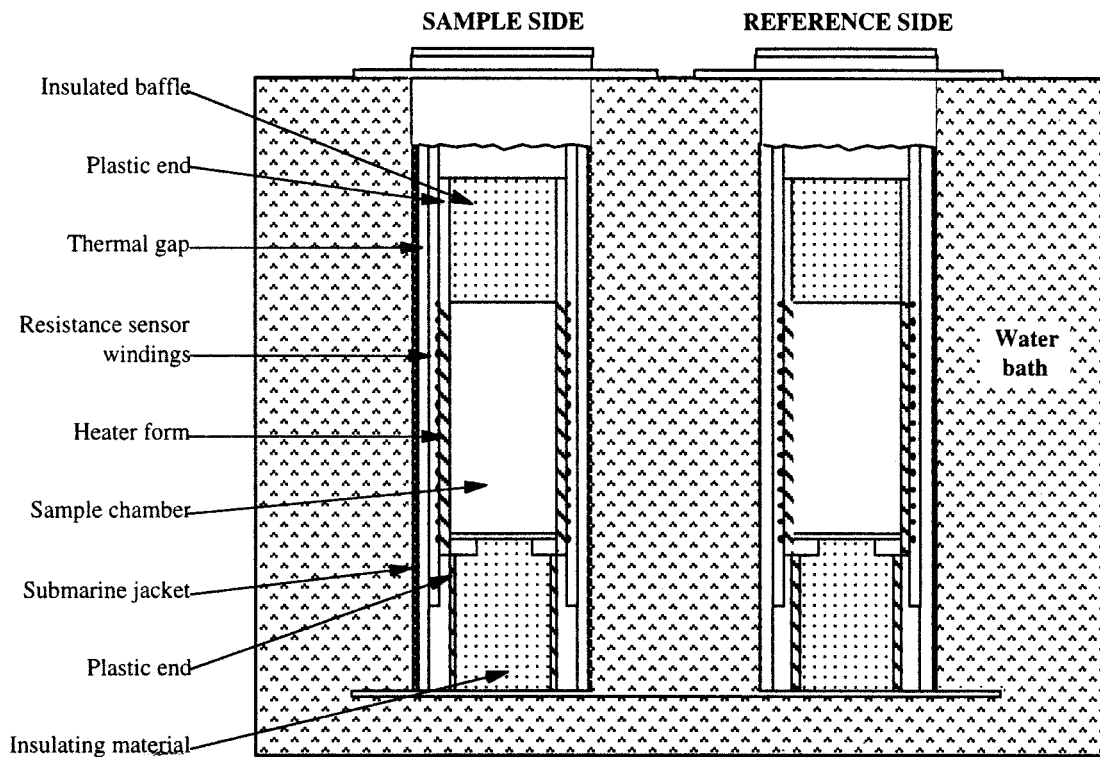


FIG. 6 Twin-Bridge Calorimeter

of the reference temperature and increase 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 machinery may increase the measurement uncertainty.

9.2.5 The base power for servo-operated calorimeters should not be less than the highest power expected from items.

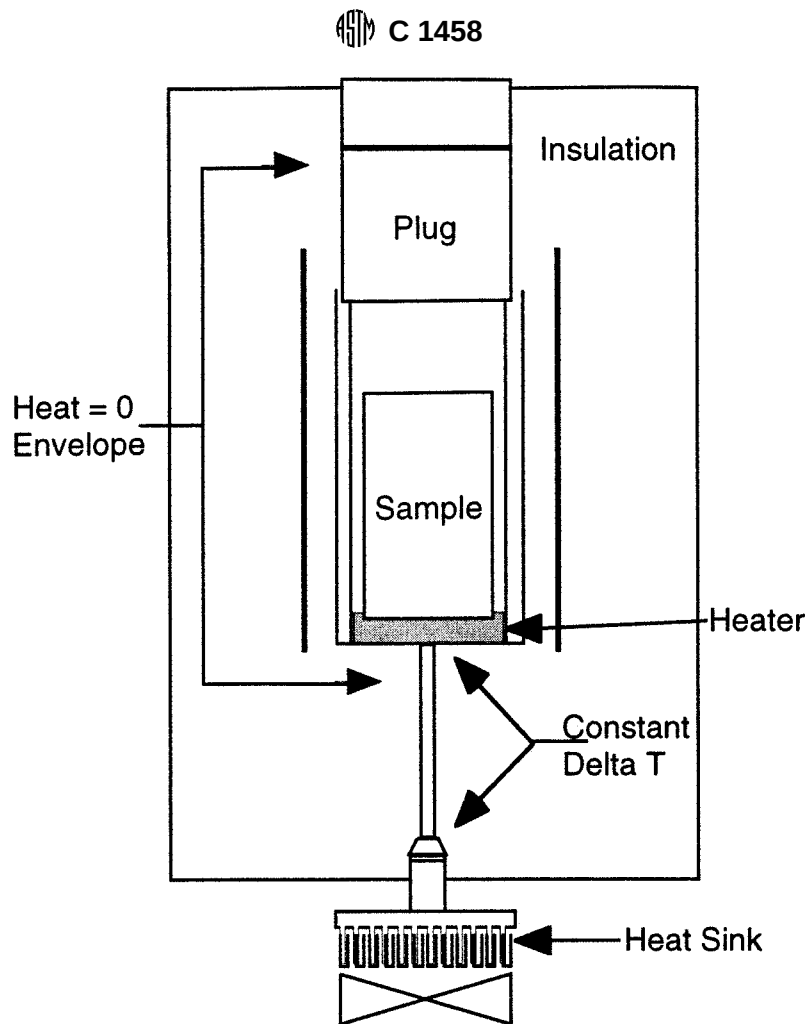


FIG. 7 Rod Calorimeter

9.2.6 Mechanical stress on the sample 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 containing small amounts of the heat-generating elements.

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 Errors can result from incorrect algorithms used for equilibrium detection or prediction. The algorithm used for equilibrium prediction that is suitable for a ^{238}Pu heat standard or electrical standard in a conductive matrix may not be suitable for items with Pu in a matrix with poor thermal conductivity.

9.2.10 Bias in the isotopic determination for plutonium will lead to a bias in the assay result.

9.2.11 The item introduced into the calorimeter should not generate enough heat such that the temperature inside the calorimeter would exceed safe limits.

9.2.12 For removable electrical standards the heater leads to the standard may serve as a heat path for heat to escape the

measurement chamber and bias the calibration.

10. Calibration Procedure

10.1 The type of calibration procedure depends on whether the calorimeter is operated in the passive or servo 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 servo mode calibration is setting the sensor output setpoint 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 should be used.

10.2.2 Initiate a baseline measurement of the bridge potential (voltage) with a calorimeter can filled with conductive material in the calorimeter chamber. There should be no heat source in the calorimeter can. Record the baseline, BP_0 (1), after equilibrium is reached.

10.2.3 Remove the calorimeter can and place the ^{238}Pu or electrical standard in the can. Center the standard in the can.

10.2.4 Close the calorimeter can and place it in the calorimeter well. For all measurements the can should be isolated thermally from room temperature by an insulating baffle.

10.2.5 Initiate the calorimeter run. Record the bridge potential 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 Repeat another baseline run. Record the baseline BP_0 (2) after equilibrium is reached.

10.2.8 Calculate the average baseline, $BP_0(av) = [BP_0(1) + BP_0(2)]/2$.

10.2.9 Use the known power generated by the ^{238}Pu heat standard or electrical standard, W_{std} , and the voltage potential generated by the standard $BP_{std} - BP_0(av)$ to calculate the calorimeter sensitivity, S .

$$S = (BP_{std} - BP_0(av))/W_{std} \quad (5)$$

10.2.9.1 The sensitivity S is not usually a constant, but varies slightly with the wattage of the standard, W_{std} , over the measurement range of the calorimeter. For one calorimeter, with an air gap thermal resistance, a standard measurement at 1 W yielded a sensitivity of 28 950 $\mu\text{V}/\text{W}$ while for another standard measurement at 11 W, it was 28 450 $\mu\text{V}/\text{W}$, a decrease in sensitivity of 1.7 %. The decreased sensitivity is qualitatively consistent with increased thermal conductivity of material forming the primary thermal resistance. For example the conductivity of dry air, a common thermal gap material, increases at the rate of 0.3 %/°C.

10.2.10 Perform a minimum of three replicate measurements at each power level using steps 10.2.1-10.2.8. This sensitivity determination at different power levels typically need only be done once after a calorimeter is installed.

10.2.11 Perform a least-squares fit of the observed sensitivities generated by the standard measurements versus W_{std} . The functional form should be as follows:

$$S = S_0 + k W_{std} \quad (6)$$

where:

S_0 = the estimated sensitivity for zero power, and

k = the slope of the varying sensitivity.

10.3 Calibration—Servo Mode:

10.3.1 Select an approximate basepower, W , at which the servo system will operate. It should be 10–20 % higher than the highest item power.

10.3.2 Determine the sensitivity S of the calorimeter by performing steps 10.2.2-10.2.9 for one ^{238}Pu or electrical standard power. Any power within the range of item powers is satisfactory.

10.3.3 Calculate the setpoint bridge potential (BP_{sp}) as follows:

$$BP_{sp} = BP_0(av) + S * W. \quad (7)$$

10.3.4 Set the controller to adjust heater power from the external power source so that BP_{sp} is maintained.

10.3.4.1 The actual supplied heater power (W_0) used to maintain BP_{sp} may be slightly different than the target value, W , because of the uncertainty in S used to calculate the setpoint.

10.3.5 Place a calorimeter can that is filled with conductive material but with no heat generating item in the calorimeter.

10.3.6 Thermally insulate the item and initiate the measure-

ment run. Once equilibrium is reached take a heater power measurement. This is the basepower W_0 corresponding to BP_{sp} .

10.3.7 Repeat the basepower measurement with no item in the sample chamber at least three times. This involves removing the calorimeter can between each of the basepower measurements at least three times and reinserting it in the calorimeter. The standard deviation of an individual basepower measurement should be less than 0.1 %.

11. Assay Procedure

11.1 *Item Measurement*—Determine the baseline bridge potential BP_0 or basepower W_0 for the calorimeter. This should be done using step 10.2.2 for the passive mode or steps 10.3.5 and 10.3.6 for the servo mode prior to the item measurement. If BP_0 or W_0 is stable the baseline or basepower measurement need not be done prior to every item measurement.

11.1.1 Load the item to be assayed into the calorimeter can, and close the can.

11.1.2 Load the can into the calorimeter measurement chamber.

11.1.3 Place the insertable insulating baffle on top of the can. Be sure the baffle top is completely down.

11.1.4 Initiate the calorimeter run.

11.1.5 When thermal equilibrium has been established or predicted the run is terminated.

11.1.6 An additional baseline (passive) or basepower (servo) run may optionally be taken after the item measurement—using step 10.2.2 or steps 10.3.5 and 10.3.6. The average of the pre- and post-item measurement baselines or basepowers may be used for BP_0 or average base power W_0 .

11.2 *Item Thermal Power Calculation*—The calculation of the item thermal power is performed differently for passive compared to servo operation.

11.2.1 *Power Calculation—Passive Mode:*

11.2.1.1 Calculate the item wattage by solving the quadratic equation for W_i as follows:

$$W_i = (BP_i - BP_0)/(S_0 + k W_i) \quad (8)$$

using the bridge potential voltage BP_i for the item, the baseline BP_0 , and the fitted parameters S_0 and k , from step 10.2.11.

11.2.1.2 The solution to the quadratic equation is, for $k < 0$, (14) as follows:

$$W_i = [-(BP_i - BP_0)/k]/Q \quad (9)$$

where:

$$Q = -0.5 \{S_0/k - [(S_0/k)^2 - 4(BP_i - BP_0)/k]^{1/2}\} \quad (10)$$

11.2.2 *Power Calculation—Servo Mode:*

11.2.2.1 Calculate the item wattage, W_i as follows:

$$W_i = W_0 - W_H \quad (11)$$

where:

W_0 = control power with no item in the calorimeter, and

W_H = control power with an item in the calorimeter.

11.2.3 If necessary, a bias correction may be made to a calorimeter measurement based on standard measurements made under a measurement control program.

11.2.4 Once the thermal power is determined the quantity of radioactive material in the container is calculated using the

equations in 4.3. For Pu, a separate isotopic analysis is required to determine P_{eff} .

12. Measurement Control Procedure

12.1 Measurement control procedures are similar for all calorimeter measurements.

12.1.1 Replicate measurements of ^{238}Pu or electrical heat standards are performed to demonstrate that the calorimeter system, hardware and software, is operating correctly. Other well-characterized nuclear materials may be used as heat standards.

12.1.2 Corrections to the thermal power of radioactive heat standards due to radioactive decay should be made.

12.1.3 Control charts formed from replicate measurements of heat standards must be used to provide quantitative means for determining that the calorimeter system is operating satisfactory prior to a single assay measurement or group of assay measurements. In addition these charts can be used to demonstrate that the calorimeter was in control during the assay runs.

12.1.3.1 Control charts may also be used to monitor baseline or basepower measurements. This can provide auxiliary information in case out-of-control operating conditions are detected.

12.1.4 Calorimeter bath temperatures can be monitored continuously to flag changes that will affect calorimeter performance. The temperatures may be evaluated using control charts or administrative limits.

12.1.5 The frequency of standards and baseline measurements are dependent on how well the calorimeter meets performance requirements and environmental conditions. Other factors to consider are size of items, range of power measurements, throughput/day, portable or permanent instruments, and data collection systems.

12.1.5.1 More details on calorimeter measurement control may be found in ANSI N15.54–1988. More general aspects of measurement control relevant to calorimetry may be found in Guide C 1009.

12.1.6 Data collected from a measurement control program can be used to calculate the precision and bias of the power measurement. A summary of the precision and bias of the power measurement obtained from replicate measurements of ^{238}Pu heat standards in production facilities over a 0.5–1.0-year period is shown in Table 1.

13. Assay Precision and Bias

13.1 Precision:

13.1.1 Calorimetric assay of plutonium requires both calorimetry and isotopic analysis, thus, the precision and bias of the assay will have components due to uncertainties in both techniques.

TABLE 1 Calorimeter Power Measurement Precision and Bias

Heat Standard Power, W	Calorimeter Diameter, m	Calorimeter Type, Operation Mode	Number of Measurements	Precision, % RSD	Bias, %
98	0.06	rod, servo	29	0.065	0.02
3.5	0.15	rod, servo	55	0.09	0.00
4.0	0.25	twin, passive ^A	22	0.05	0.03
4.9	0.30	twin, passive ^A	34	0.06	0.05

^APooled results from two calorimeters.

13.1.2 The precision of a calorimeter measurement is dependent on the sensitivity of the calorimeter, baseline stability, and item power.

13.1.3 A list of major factors that can affect the precision of the gamma-ray isotopic assay and mass spectrometric modes of isotopic analysis are described in Test Methods C 1030 and C 697, respectively.

13.1.4 *Plutonium in $\text{PuO}_2\text{-UO}_2$ Mixed Oxide*—Generally, the greater the thermal power in a calorimeter, the better the precision. To illustrate this the relative precision observed from repetitive calorimeter measurements of six items containing 26 to 258 g of Pu (17 % ^{240}Pu) in $\text{PuO}_2\text{-UO}_2$ (26 % Pu) was calculated and the results are shown in Table 2. These measurements were made over a 56-day period with a water bath twin-bridge over-under calorimeter. The items were loaded robotically allowing for continuous operation. A common P_{eff} factor for all six items was determined using the plutonium isotopic composition and ^{241}Am content that was determined by mass spectrometry and alpha counting. The calorimeter can size was 0.06-m diameter $\times$ 0.16-m high. The calorimeter measurement time was fixed at 1 h. The calorimeter was run in the servo mode and the items were preconditioned to reduce measurement time.

13.1.5 *Plutonium in PuO_2* —The results of multiple calorimetric and gamma-ray isotopic measurements by three facilities on identical standards each containing 400 g of well-characterized Pu (6 % ^{240}Pu) in PuO_2 are shown in Table 3. The Pu content and isotopic composition used as reference values of the mother lot of PuO_2 material used for these standards were measured by coulometry and mass spectrometry/alpha counting by four analytical laboratories. The calorimetry and gamma-ray measurements used to determine Pu mass variabilities and biases reported in Table 3 were taken over a one-year period. The within-facility variability and the bias of the calorimetric/gamma-ray assay were calculated from results reported by each facility decayed to a common date. Each facility used different gamma-ray analysis codes for the isotopic measurements. For some the reported values are the averages of measurements of the standard item with different calorimeters.

TABLE 2 Calorimetry/Mass Spectrometry Measurements of Plutonium in $\text{PuO}_2\text{-UO}_2$ Mixed Oxide^{A,B}

ID	Mass ^{B,C} , g by Chem	Mass, g ^C , by Cal ^D /MS/ α ^E	Precision, g 1 SD	Precision %RSD ^F	Bias, g	Bias, %
4	257.70	257.54	0.14	0.06	−0.16	−0.06
5	206.09	206.06	0.13	0.06	−0.03	−0.02
6	206.18	206.12	0.14	0.07	−0.06	−0.03
7	128.81	128.94	0.12	0.09	0.13	0.10
8	77.28	77.35	0.12	0.15	0.07	0.09
9	25.79	25.99	0.11	0.42	0.20	0.78

^A Unpublished results.

^B Mass of plutonium determined by coulometry using reference material NBS 949E. Pu percentage of mixed oxide, 0.25759, based on triplicate measurements of six samples.

^C Pu masses reported here decayed to a common date.

^D Final results based on 117 replicate calorimeter measurements/item.

^E Isotopic composition determined by 12 replicate measurements by mass spectrometry (^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu) and six replicate alpha counting (^{238}Pu , ^{241}Am) measurements. Average Pu isotopic and ^{241}Am results were used to calculate P_{eff} used for all calorimeter measurements.

^F Precision due to calorimetry power replicate measurements.

TABLE 3 Calorimetry/Gamma-Ray Assay Measurement of 400 g of Pu in PuO₂^A (15)

Facility	Within-Facility Variability, g	Within-Facility Variability, %RSD	Bias, g	Bias, %
A ^B	1.5	0.38	0.03	0.01
B ^B	1.5	0.38	-0.40	-0.10
C ^C	1.4	0.36	0.04	0.01

^AAll masses are in grams of plutonium decayed to a common date.

^BUsed multiple water bath twin-bridge calorimeters.

^CUsed "air-bath" calorimeter.

13.2 Bias:

13.2.1 The bias of calorimetric assay can be determined by the measurement of certified reference materials or well-characterized items with known elemental and isotopic compositions.

13.2.2 Plutonium in PuO₂-UO₂ and PuO₂ (15). Biases for calorimetric assay of Pu in PuO₂-UO₂ mixed-oxide powder and Pu in PuO₂ powder are shown in Tables 2 and 3, respectively. This data shows that the biases for this measurement application are typically less than 0.1 %.

13.2.3 *Plutonium in Salt Residues (16,17)*—In two separate studies calorimetry and gamma-ray spectroscopy measurements were used to assay items containing a mixture of KCl, MgCl₂, and NaCl residues containing PuCl₃ and AmCl₃, as well as, shards of MgO crucibles and plutonium metal shot. The material in each item was sampled and analyzed for Pu. The average relative biases between the calorimetry assay and alternative assay is shown in Table 4. A twin bridge water bath calorimeter was used for the thermal power measurements in both studies.

13.2.4 *Americium in Salt Residues (16,17)*—The same mea-

TABLE 4 Bias—Calorimetry/Isotopic Measurements of Pu and Am in Molten Salt Residues

Mass Range, g Pu	No. of Items	Mass Range, g Am	Bias, % Pu	Bias, % Am	References
35–416	10	0.4–18	1.5 ± 0.5 ^A	0.2 ± 0.4 ^A	16
214–414	9	5–26	1.6 ± 0.6 ^B	0.2 ± 0.6 ^B	17

^A*Chemistry*—Pu and Am elemental analysis by Isotopic Dilution Mass Spectrometry. Aliquots taken of blended salt matrix for analysis. Pieces of Pu metal removed, oxidized, and returned to matrix before blending.

Calorimetry—Isotopic analysis for P_{eff} determination by gamma-ray spectroscopy.

^B*Chemistry*—Entire matrix of each item dissolved and liquid samples measured by x-ray fluorescence for Pu and gamma counting for Am.

Calorimetry—Isotopic analysis for P_{eff} determination by gamma-ray spectroscopy and facility stream averages of certain Pu isotopic ratios.

surement techniques described in section 13.2.3 were used to assay the ²⁴¹Am content of the items containing molten salt residues, as well as, the Pu content. Gamma-ray spectroscopy was used to determine the ²⁴¹Am/Pu ratio simultaneously with plutonium isotopic ratios. The biases are shown in Table 4.

13.2.5 *Tritium (18)*—Calorimetry was used to measure the quantity of tritium gas in containers. Since tritium was the only radioactive isotope, no isotopic measurements were required for the assays. After the calorimeter measurement the gas was quantitatively transferred to tanks with calibrated volumes, and the quantity of tritium was determined using calibrated pressure and temperature transducers and mass spectrometric analyses. A comparison of measurement results between calorimetry and PVT,MS was made for 50 containers. The tritium content of the containers ranged from 15 to 16 g. The relative mean bias for the calorimeter assay compared to PVT,MS was -0.12 ± 0.05 % (RSD_{mean}). A twin bridge water bath calorimeter was used for the calorimeter measurements.

APPENDIXES

(Nonmandatory Information)

X1. MEASUREMENT OF PLUTONIUM CONTENT

X1.1 The amount of plutonium in an item can be calculated from the thermal power measured by calorimetry (W) and from the item effective specific power (W/g) as shown in Eq X1.1:

$$m = \frac{W_i}{P_{\text{eff}}} \quad (\text{X1.1})$$

X1.1.1 The effective specific power of the material in the item is calculated in Eq X1.2:

$$P_{\text{eff}} = \sum_i R_i P_i \quad (\text{X1.2})$$

where:

R_i = abundance of the i -th isotope ($i = {}^{238}\text{Pu}, {}^{239}\text{Pu}, {}^{240}\text{Pu}, {}^{241}\text{Pu}, {}^{242}\text{Pu}, \text{ and } {}^{241}\text{Am}$) of the Pu in the item expressed as a weight fraction, g isotope/g-Pu, and P_i = a physical constant, the specific power of the i -th isotope in the item in W/g.

X1.1.2 The isotopic fractions, R_i , can be determined destructively by mass spectrometry and alpha spectrometry, or nondestructively by gamma-ray spectrometry. Liquid scintilla-

tion counting may be used as well as alpha spectrometry for determination of the ²⁴¹Am abundance by means of destructive analysis. The terms $R_i P_i$ are the contribution of each isotope to the total item power. The isotopic fractions R_i are not constant, but change as the constituent isotopes in the item decay or grow in.

X1.2 The isotopic specific powers, P_i , for some Pu isotopes have been directly measured or can be derived from Eq X1.3. The specific powers and half-lives for Pu isotopes and ²⁴¹Am as listed in ANSI N15.22–1987 are listed in Table X1.1. The half-lives were established after a data review of collaborative and individual experiments by the USDOE Half-Life Evaluation Committee. The half-lives and specific powers of ²³⁹Pu and ²⁴⁰Pu were determined from collaborative experiments on nearly pure isotopic samples. The specific powers of ²⁴¹Pu and ²⁴²Pu were measured by individual experimenters and the specific power of ²³⁸Pu and ²⁴¹Am were determined using Eq X1.3:

TABLE X1.1 Nuclear Decay Parameters for Pu Calorimetric Assay^A

Isotope	Half-Life, Years	Standard Deviation, Years	Specific Power, W/g	Standard Deviation, W/g	References
²³⁸ Pu	87.74	0.04 (0.05 %)	0.56757	0.00026 (0.05 %)	(19,20)
²³⁹ Pu	24 119	16 (0.11 %)	1.9288×10^{-3}	0.0003×10^{-3} (0.02 %)	(20-22)
²⁴⁰ Pu	6564	11 (0.17 %)	7.0824×10^{-3}	0.0020×10^{-3} (0.03 %)	(23-28)
²⁴¹ Pu	14.348	0.022 (0.15 %)	3.412×10^{-3}	0.002×10^{-3} (0.06 %)	(29-33)
²⁴² Pu	376 300	900 (0.24 %)	0.1159×10^{-3}	0.00026×10^{-3} (0.22 %)	(34)
²⁴¹ Am	433.6	1.4 (0.32 %)	0.1142	0.00042 (0.37 %)	(32,35)

^ANumbers in parentheses are % relative standard deviation (RSD).

$$P_i = \frac{2119.3}{T_{1/2(i)} A_i} Q_i \quad (X1.3) \quad A_i = \text{gram atomic weight of isotope } i.$$

where:

Q_i = total disintegration energy (MeV) for alpha-particle emitters, or the average energy (MeV) of beta particle emitters;

$T_{1/2(i)}$ = half-life (years) of isotope i ; and

X2. PROPAGATION OF ERROR FOR PLUTONIUM AND ²⁴¹AM MASS

X2.1 In Eq X1.1, the measurement of the thermal power, W , is independent of the measurement of the effective specific power, P_{eff} , and thus, the relative uncertainty for the plutonium mass, M , can be written as follows:

$$\frac{\sigma_M}{M} = \left[\frac{\sigma_W^2}{W^2} + \frac{\sigma_{P_{eff}}^2}{P_{eff}^2} \right]^{1/2} \quad (X2.1)$$

X2.1.1 The uncertainty in the power measurement, σ_W , can be obtained from replicate power measurements of heat standards or from historical data. It should include both precision and bias components. The uncertainty in P_{eff} , $\sigma_{P_{eff}}$, comes from the uncertainty in the isotopic fractions, R_i , and isotopic specific powers, P_i . The uncertainties in the isotopic fractions are determined from uncertainties in the various techniques that might be used for the isotopic analysis, such as mass spectroscopy, alpha counting, or gamma-ray spectroscopy. There are sufficient gamma-rays in plutonium to provide independent measured isotopic ratios of the major contributors to the item thermal power: ²³⁸Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Pu, and ²⁴¹Am with respect to ²³⁹Pu that allow R_i to be calculated. Ref (36) discusses this in more detail. The uncertainties in the isotopic specific powers, P_i , as determined by different experiments, are given in Table X1.1. The test method for determining isotopic

composition by gamma-ray spectroscopy is described in Test Method C 1030. Several gamma-ray analysis codes are available commercially that not only provide the isotopic composition, but also the uncertainties of the isotopic fractions and the specific power of the item being measured. Error propagation of the isotopic fractions is discussed in Ref (36).

X2.1.2 The uncertainty of the ²⁴¹Am mass mixed with Pu is as follows:

$$\frac{\sigma_M}{M} = \left\{ \left[\left(\frac{\sigma_W}{W} \right)^2 + \left(\frac{K}{P_{eff}} \right)^2 \left[\left(\frac{\sigma_K}{K} \right)^2 + \left(\frac{\sigma_{R_{Am}}}{R_{Am}} \right)^2 \right] + \left(\frac{R_{Am} \sigma_{P_{Am}}}{P_{eff}} \right)^2 \right] \right\}^{1/2} \quad (X2.2)$$

where:

$$K = P_{eff} - P_{Am} R_{Am} \quad (X2.3)$$

where:

P_{Am} = the specific power of ²⁴¹Am,
 R_{Am} = the mass ratio of ²⁴¹Am to plutonium,
 W = the thermal power,
 σ_W = the thermal power uncertainty,
 $\sigma_{R_{Am}}$ = the uncertainty in the ²⁴¹Am mass ratio, and
 $\sigma_{P_{Am}}$ = the uncertainty of the ²⁴¹Am specific power.

X3. PROPAGATION OF ERROR FOR TRITIUM MASS

X3.1 The uncertainty of a calorimetric assay of tritium can be calculated using Eq X2.1. The uncertainty of the effective specific power, P_{eff} , of tritium is the same as the isotopic specific power, 0.00045 W obtained from 4.3.1. Dividing by the specific power of tritium, 0.3240 W/g, results in the following:

$$\frac{\sigma_{P_{eff}}}{P_{eff}} = 0.0014 \quad (X3.1)$$

So for tritium the relative uncertainty of the tritium mass is as follows:

$$\frac{\sigma_{\text{tritium}}}{M_{\text{tritium}}} = \left[\left(\frac{\sigma_W}{W} \right)^2 + (0.0014)^2 \right]^{1/2} \quad (X3.2)$$

REFERENCES

- (1) "Tritium Decay Energy," W. L. Pillinger, J. J. Hentges, and J.A. Blair, *Phys. Rev.*, 1961, pp. 232–233.
- (2) "Handbook of Nuclear Safeguards Measurements," edited by D. R. Rogers, NUREG/CR-2078, 1981, pp. 533–550.
- (3) "Passive Non-Destructive Assay of Nuclear Materials," edited by Doug Reilly, Norbert Ensslin, and Hastings Smith, Jr., NUREG/CR-5550, 1991, pp. 641–660.
- (4) "The Use of Calorimetry for Plutonium Assay," J. A. Mason, UKAEA Report SRDP-R100, 1982.
- (5) "In-Field Calibration of Neutron Correlation Counter via Calorimetry and High Count Rate Gamma-Ray Isotopic Abundance Measurements," IAEA-SM-293/126, *Nuclear Safeguards Technology*, 1986, Vol 2, IAEA, 1987, pp. 239–249.
- (6) "In Field Calibration of a Neutron Correlation Counter Via Calorimetric Assay," 2nd International Workshop on Calorimetric Assay, Santa Fe, New Mexico, 27 October 1994.
- (7) "Selection of Nondestructive Assay Methods: Neutron Coincidence or Calorimetric Assay?" T. Cremers and J. R. Wachter, *Nuclear Materials Management*, Vol XIII, p. 483–486, Proceedings Issue, 1994.
- (8) "Test and Evaluation of the Argonne BPAC10 Series Air Chamber Calorimeter Designed for 20 Minute Measurements," R. B. Perry, S. Fiarman, E. A. Jung, and T. Cremers, ANL-90/36, 1990.
- (9) "Heat Standards Calibration in the DOE Complex," L. Carrillo, C. R. Rudy, S. M. Long, J. McDaniel, and W. W. Rodenburg, *Nuclear Materials Management*, Vol XXVI Proceedings Issue, 1997.
- (10) "High Wattage Comparison of the MECH Probe and ^{238}Pu Heat Standards at Los Alamos," D. P. Renz, J. R. Wetzel, M. B. Severinghaus, R. I. Sheldon, and L. Bruchner, *Nuclear Materials Management*, Vol XXIII, Proceedings Issue, 1994, pp. 95.
- (11) "Electrical and Heat Standard Calibration of Calorimeters for Plutonium and Tritium Measurement," J. A. Mason and M. I. Thornton, Proceedings of the 17th ESARDA Symposium on Safeguards and Nuclear Materials Management, Aachen, May 1995, pp. 581–583.
- (12) "High Precision Large Sample Plutonium Calorimetry Measurement," J.A. Mason, J. G. Fleissner, J. L. Valdez, G. Campbell, and J. West, *Nuclear Materials Management*, Vol XXVI Proceedings Issue, 1997.
- (13) "Dry Calorimetry at the Savannah River Site," L. A. Refalo, Proceedings of the 1st International Workshop on Calorimetry, Ispra, Italy, 1992, pp. 39–41.
- (14) "Numerical Recipes," W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, Cambridge University Press, 1986.
- (15) "Calorimetry Exchange Program, Quarterly/Annual Data Report, Calendar Year 1998," prepared by M. Irene Spaletto and David T. Baran, NBL-353, May 1999.
- (16) "Isotopic Ratios and Effective Power Determined by Gamma-Ray Spectroscopy vs. Mass Spectroscopy for Molten Salt Extraction Residues," V. L. Longmire, T. L. Cremers, W. A. Sedlacek, S. M. Long, A.M. Scarborough, and J. R. Hurd, *Nuclear Materials Management*, Vol XXXI Proceedings Issue, 1990.
- (17) "Comparison of Destructive and Nondestructive Assay of Heterogeneous Salt Residues," J. G. Fleissner and M. W. Hume, RFP-3876, 1986.
- (18) "A Measurement Control Study for Tritium Gas," C. N. Lindsay, R. E. Sprague, and J. A. Brandenburg, MLM-2441, (1987).
- (19) Wapstra, A. H. and Bos K., The 1977 Atomic Mass Evaluation, *Atomic Data and Nuclear Data Tables*, Vol 19, No. 3, 1977.
- (20) Strohm, W. W., "The Measurement of the Half-Life of Plutonium-239 by the U. S. Half-Life Evaluation Committee, *International Journal of Applied Radiation and Isotopes*, Vol 29, 1978, pp. 481–483.
- (21) Seabaugh, P. W. and Jordan, K. C., "Calorimetric Determination of the Half-Life of Plutonium-239," *International Journal of Applied Radiation and Isotopes*, Vol 29, 1978, pp. 489–496.
- (22) Gunn, S. R., "A Calorimetric Determination of the Specific Power of Plutonium-239, *International Journal of Applied Radiation and Isotopes*, Vol 29, 1978, pp. 497–499.
- (23) Rudy, C. R., Jordan, K. C., and Tsugawa, R., "Calorimetric Determination of the Half-Life of Plutonium-240," *International Journal of Applied Radiation and Isotopes*, Vol 35, 1984, pp. 177–180.
- (24) Lucas, L. L., and Noyce, J. R., "The Half-Life of Plutonium-240," *International Journal of Applied Radiation and Isotopes*, Vol 35, 1984, pp. 173–176.
- (25) Beckman, R. J., Marsh, S. F. Abernathy, R. M., and Rein, J. E., "Plutonium-240 Half-Life Determined by Isotope-Dilution Mass Spectrometric Measurement of the Grown-in Uranium-236," *International Journal of Applied Radiation and Isotopes*, Vol 35, 1984, pp. 163–169.
- (26) Steinkruger, F. J. Matlack, G. M., Beckman, R.T., "The Half-Life of ^{240}Pu Determined by Specific-Activity Measurements," *International Journal of Applied Radiation and Isotopes*, Vol 35, 1984, pp. 171–172.
- (27) Jaffey, A. H., Diamond, H., Bentley, W. C., Graczyk, D. G., Flynn, K. F., "Half-Life of ^{240}Pu , *Physical Review, C*, Vol 18, 1978, pp. 969–974.
- (28) Strohm, W. W., "The Measurement of the Half-Life of Plutonium-240 by the U. S. Half-Life Evaluation Committee," *International Journal of Applied Radiation and Isotopes*, Vol 35, 1984, pp. 155–157.
- (29) Marsh, S. F., et al, "Plutonium-241 Half-Life Determined by Mass Spectrometric Measurements of ^{241}Pu : ^{242}Pu Ratios," *International Journal of Applied Radiation and Isotopes*, Vol 31, 1980, pp. 629–631.
- (30) Garner, E. L., Machlan, L. A., "Mass Spectrometric Measurements to Determine the Half-Life of ^{241}Pu , NBS Special Publication 582, National Bureau of Standards, 1980, June, pp. 34–40.
- (31) DeBievre, P., "Isotope Dilution Mass Spectrometry; Some Present Limitations and Possibilities to Overcome These," Minutes of SALE program participants meeting, 1981 July 8–9 pp. 188–207.
- (32) Jordan, K. C., unpublished results, Monsanto Research Corporation, Mound Laboratory, Miamisburg, OH (1982).
- (33) Oetting, F. L., "Average Beta Energy of Plutonium-241 by Calorimetry, *Physical Review*, Vol 168, No. 4, 1968, pp. 1398–1401.
- (34) Osborne, D. W., Flotnow, H. E., "Half-Life of ^{242}Pu from Precise Low Temperature Heat Capacity Measurements," *Physical Review C*, Vol 14, No. 3, 1976, pp. 1174–1178.
- (35) Oetting, F. L., Gunn, S. R., A Calorimetric Determination of the Specific Power and half-life of ^{241}Am , *Journal of Inorganic and Nuclear Chemistry*, Vol 29, 1967, pp. 2659–2664.
- (36) Sampson, T. E., Gunnink, R. "The Propagation of Errors in the Measurement of Plutonium Isotopic Composition by Gamma-Ray Spectroscopy," *Journal of the Institute of Nuclear Materials Management*, Vol XII, No 2. 1983 Summer, pp. 39–48.

 C 1458

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