
Calorimetric Assay Instruments

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22.1 INTRODUCTION

This chapter describes some modern calorimeter systems currently in use throughout the nuclear fuel cycle for the nondestructive assay of plutonium. Since 1950, Mound Laboratory (Monsanto Research Corporation, Miamisburg, Ohio) has built over 200 calorimeters of various designs for nuclear material control and accountability, primarily for US Department of Energy (DOE) facilities. Occasionally a few DOE and commercial nuclear facilities have built their own calorimeters. Argonne National Laboratory (University of Chicago, Argonne, Illinois) has built several for use by the International Atomic Energy Agency (IAEA). The calorimeters described in this chapter represent the types of instruments in use today; they include those built by Mound, Argonne, Rocky Flats, and General Electric (GE).

The Mound calorimeters described in this chapter are the analytical calorimeter, the transportable calorimeter, the twin-bridge production calorimeter, and the gradient bridge calorimeter; the Argonne calorimeters are the small-sample calorimeter, the bulk assay calorimeter, and the one-meter fuel rod calorimeter. Also described are Rocky Flats production calorimeters and the GE irradiated fuel assembly calorimeter. The examples given in this chapter are preceded by a summary of typical calorimeter system components and design considerations.

22.1.1 Calorimetric Assay System Components

A typical modern system designed for the calorimetric assay of plutonium-bearing materials consists of the following components:

(a) A precisely machined heat flow calorimeter body of the twin-bridge, over/under bridge, or gradient bridge type. Each cell includes an electric heater circuit for accurate duplication of sample power.

(b) A bridge circuit for the precise measurement of temperature differences between the sample chamber and the environmental heat sink. The electric circuitry includes high-quality potentiometers and digital voltmeters.

(c) A digital or analog readout device, which is usually a minicomputer. The computer controls calorimeter operation, data collection and storage, and data analysis.

(d) Electrical or radioactive heat source calibration standards. Electrical measurements are used to determine temperature differences and to control electrical heater circuits for sample power duplication. Alternatively, radioactive heat source standards may be available to the system for calibration and power determination.

22.1.2 Calorimeter Design Considerations

In the design of a calorimetric assay instrument, many important factors dictate the appearance of the final product. It is not possible to design a single, universal calorimeter that is applicable to all measurement situations. This section lists some of the factors that influence calorimeter design.

(a) *Sample size.* The physical size of the sample dictates the dimensions of the sample chamber; tight thermal coupling of the sample to the calorimeter is essential to minimize assay time. The diameters of sample chambers on existing calorimeters range from 1 to 30 cm.

(b) *Sample power.* In general, high-power samples require low-sensitivity calorimeters with low thermal resistance, and low-power samples require sensitive microcalorimeters with high thermal resistance.

(c) *Calibration.* Calorimeter design is a function of the calibration method to be used—radioactive heat standards or electrical standards.

(d) *Construction materials.* The heat capacities and thermal conductivities of the materials used in construction influence performance.

(e) *Throughput.* The available time per assay influences the choice of calorimeter type and method of operation and the number of calorimeters needed.

(f) *Accuracy.* The desired assay accuracy must be weighed against the needed throughput and available space in choosing the type of calorimeter and the method of operation.

(g) *Plant environment.* Calorimeter design is affected by the working environment and the available floor space. An in-line production plant environment requires different design considerations than a laboratory setting.

22.2 SMALL CALORIMETERS FOR LABORATORY SAMPLES

Small calorimeters can be used in analytical chemistry laboratories to provide nondestructive assays of small plutonium samples. They can be used to determine effective specific power by the empirical method, in which a small sample of plutonium is assayed nondestructively by calorimetry and then destructively by chemistry. Also, small calorimeters provide a means for evaluating sampling errors.

22.2.1 Mound Analytical Calorimeter

The Mound analytical calorimeter was developed specifically for use in an analytical chemistry laboratory (Ref. 1). It is a compact instrument (Figure 22.1) designed for sample sizes up to 5 cm³ (about 10 g of plutonium). The calorimeter includes an automatic sample loader with a pre-equilibrium position and is designed to fit inside a standard 3-ft glovebox. For samples producing 10 mW of power, the precision and accuracy are better than 0.1%. Performance data for the analytical calorimeter are summarized in Table 22-1 (Refs. 1 and 2). The gram values for accuracy, precision, and range are for low-burnup plutonium with an effective specific power of 2.3 mW/g and with well-known isotopic composition.

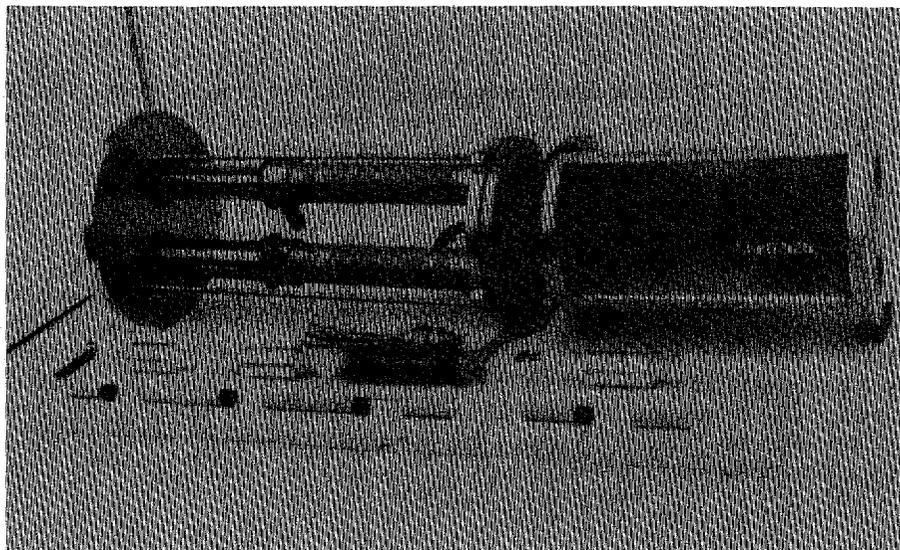


Fig. 22.1 View of a disassembled Mound analytical calorimeter for the assay of small plutonium samples in 2-dram glass vials. (Photograph courtesy of Mound Laboratory.)

Table 22-1. Performance data for the Mound analytical calorimeter, assuming 2.3 mW/g and known isotopics (Refs. 1 and 2)

Accuracy	0.008 g
Precision	0.008 g
Range	0.4 to 10 g (1 to 23 mW)
Measurement Sensitivity	0.4 g or 1 mW in a 5-cm ³ container
Assay Time	30-100 min

A wide variety of plutonium-bearing materials, including metal, oxide, and mixed-oxide, have been measured in the analytical calorimeter and then dissolved and analyzed for plutonium content. Comparisons of the results demonstrated that calorimetric assay is both accurate and precise.

22.2.2 Argonne Small-Sample Calorimeter

The Argonne small-sample calorimeter is designed for the assay of up to 6 cm³ of mixed-oxide fuel in the form of pellets, powders, and solutions (Refs. 3 and 4). Its maximum power range is 45 mW, which is equivalent to 20 g of low-burnup plutonium. The unique feature of this system is its portability, which makes it useful for on-site inspections in the field. Figure 22.2 shows the two modules that make up the system: the isothermal calorimeter and the data acquisition module. The total weight is 18 kg.

The small-sample calorimeter is an isothermal gradient bridge calorimeter. It is a "dry" or "air chamber" calorimeter because of the absence of a water bath heat sink, as described in Section 21.5.4 and shown in Figure 21.7 of Chapter 21. This design requires operation in the constant temperature servo-control mode to obtain good accuracy. Performance data for the small-sample calorimeter are given in Table 22-2 (Refs. 4 through 6). The calorimeter has a precision of about 0.1% in 20 min (Ref. 4). The short assay time and good precision are partly due to double encapsulation of the samples in metal containers to maximize heat transfer.

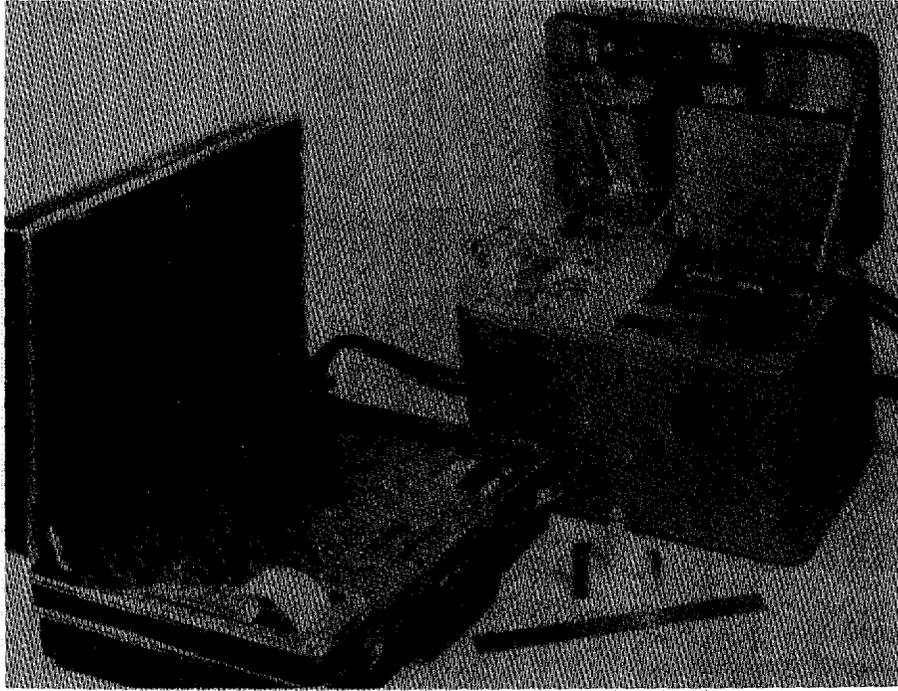


Fig. 22.2 Argonne small-sample calorimeter (right) and its data acquisition system (left). (Photograph courtesy of Argonne National Laboratory.)

Table 22-2. Performance data for the Argonne small-sample calorimeter, assuming 2.3 mW/g and known isotopics (Refs. 4 through 6)

Accuracy	0.01 g
Precision	0.01 g
Range	1 to 20 g (2 to 45 mW)
Measurement Sensitivity	1 g
Assay Time	20 min

The data acquisition module automatically performs electrical calibrations over a selected input range. The calibrations are performed with the aid of a microprocessor calculating the reference voltage to be applied across the calibration resistor coil. This electrical calibration simulates a set of plutonium standards over the measurement range of the instrument (Ref. 4).

An independent test and evaluation of the calorimeter (Ref. 5) found insensitivity to operator and environmental effects. However, the study recommended preheating the sample capsules to avoid a shift in the reference-power baseline. The calorimeter was calibrated with plutonium heat standards, and use of an electrically heated capsule was recommended to check the calibration.

22.3 TRANSPORTABLE CALORIMETERS

Some calorimeters have been designed to be transportable for use at different nuclear fuel-cycle facilities during periodic inventory verification exercises. This portability provides the nuclear material inspector with the ability to verify inventory items independently. In addition, a gamma-ray spectrometer is used to determine plutonium isotopic concentrations so that the calorimeter power measurement can be converted to plutonium mass.

22.3.1 Mound Transportable Calorimeter

The Mound transportable calorimeter was developed for use by DOE inspectors (Refs. 1 and 7). It is similar in design to the larger in-plant instrument described in Section 22.4.1 below, except that the constant temperature bath is replaced by a heat exchanger, a pump, and a small temperature-controlled reservoir. Water from the small bath is pumped through the heat exchanger to maintain the calorimeter at constant temperature. This change significantly reduces the size and weight of the system. The calorimeter is an over/under bridge type (Ref. 1).

Figure 22.3 shows the small water reservoir, the electronics, and the calorimeter body mounted on an aluminum cart. Because the total weight of the system is only 200 kg, it can be moved by one person. The sample chamber can accommodate samples up to 13 cm in diameter and 20 cm high, with a power range of 0.5 to 10 W. The data acquisition system reads the bridge potential, monitors the water reservoir to within 0.001°C, and uses the end-point prediction technique to reduce assay time (Ref. 7).

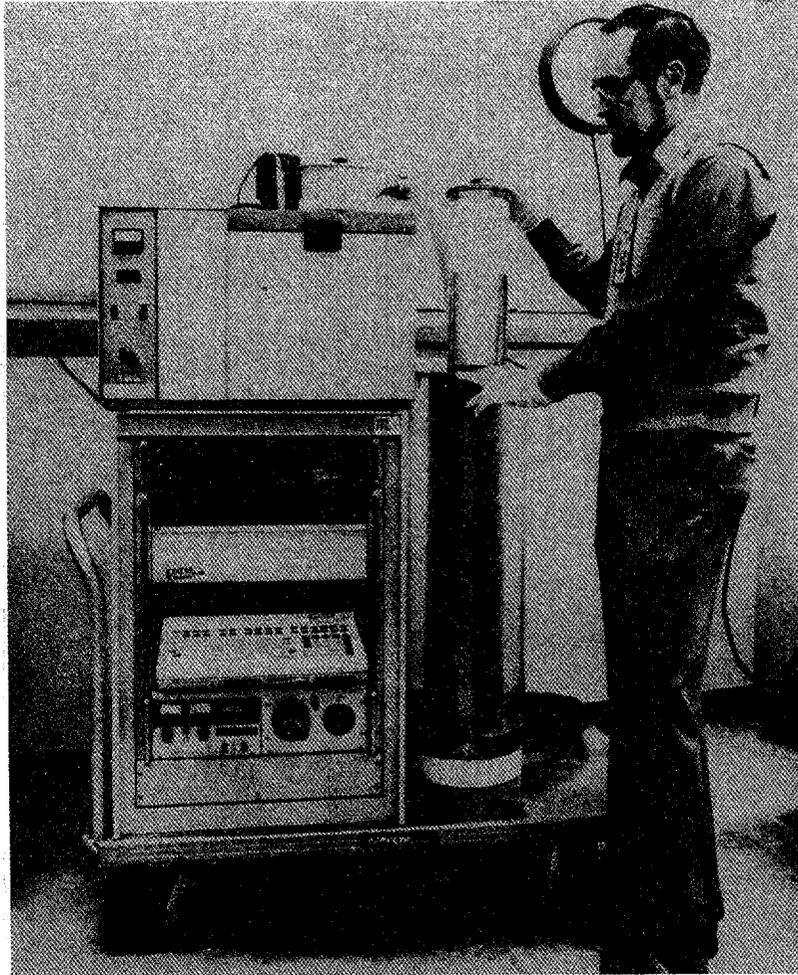


Fig. 22.3 Mound transportable calorimeter system showing water bath temperature controller (upper left), electronics and data acquisition system (lower left), and over/under bridge calorimeter (right). (Photograph courtesy of Mound Laboratory.)

In the field, about 4 h are required after unpacking for the water reservoir to come under temperature control and for the intrinsic germanium gamma-ray detector to cool down. During operation, about 4 h are required for each calorimeter power measurement and each gamma-ray spectrum measurement. An on-line computer is used to obtain the isotopic composition from the gamma-ray spectra and to calculate the effective specific power. Reported measurement uncertainties are about 0.3% for the power determination, including precision error, calibration error, and heater lead error, and 1 to 2% for the gamma-ray specific power determination (Ref. 6). Performance data for the transportable calorimeter are summarized in Table 22-3 (Ref. 7).

Table 22-3. Performance data for the Mound transportable calorimeter, assuming 2.3 mW/g and known isotopics (Ref. 7)

Accuracy	0.3%
Precision	0.1%
Range	200 g to 4.4 kg (0.5 to 10 W)
Assay Time	4 h

22.3.2 Argonne Bulk Assay Calorimeter

The Argonne bulk assay calorimeter (Figure 22.4) was developed for use by IAEA inspectors (Refs. 6 and 8). It is designed to measure sealed canisters holding up to 3 kg of high-burnup recycle plutonium in the form of metal, powder, or scrap. This isothermal gradient air calorimeter (see Section 21.5.4 of Chapter 21) operates in the servo-control mode (see Section 21.6.3 of Chapter 21). It consists of five nested servo-controlled cylinders separated from each other by heat-conducting epoxy. The system includes sample preheaters and is completely microprocessor controlled. The sample chamber can accept cans 11 cm in diameter and 33 cm high.

Performance data (Refs. 6, 8, 9, and 10) for the bulk assay calorimeter are summarized in Table 22-4. In one experiment, 18-cm-long mixed-oxide fuel rods were assayed in different numbers and different arrangements (Ref. 6). The assays were unaffected by the geometrical arrangement, and the response per gram was constant to within 0.1% as the number of rods was varied. This behavior differs from that of other assay techniques, where gamma-ray self-attenuation or neutron multiplication may result in nonlinear effects.

A field test of the bulk assay calorimeter was conducted at a mixed-oxide fuel fabrication plant in Belgium (Ref. 9). Five plutonium oxide cans were assayed to an average accuracy of 0.2% (1σ). Most plutonium cans at the facility had a diameter of more than 11 cm and could not be assayed during this test. A recent evaluation of the bulk assay calorimeter at three United Kingdom Atomic Energy Agency facilities involved the measurement of more than 70 standards consisting of plutonium oxide, metal, fuel pellets, or mixed oxide (Ref. 10). Using calorimeter power values and gamma-ray isotopic values, the ratio of measured to declared plutonium mass typically differed from 1 by 0.3%. The absence of a water jacket was considered helpful for such field measurements.

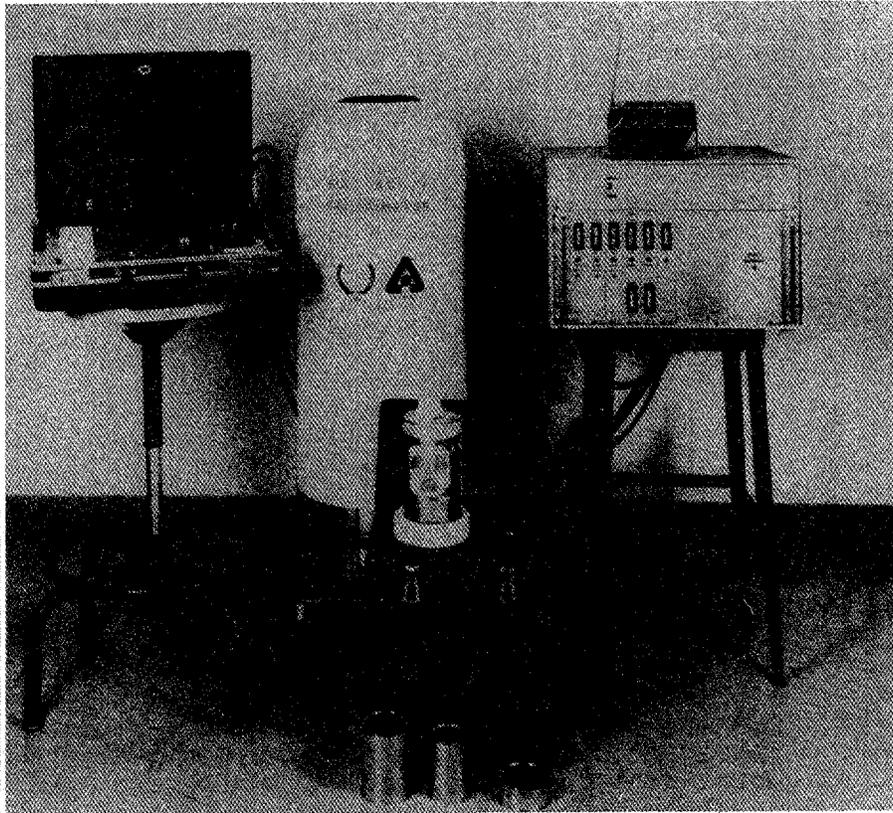


Fig. 22.4 Argonne bulk assay calorimeter (center), with data acquisition system (left), control circuit power bin (right), and sample preheating chambers and calorimeter canisters (front). (Photograph courtesy of Argonne National Laboratory.)

Table 22-4. Performance data for the Argonne bulk assay calorimeter (Refs. 6, 8, 9, and 10)

Accuracy	0.1 to 0.9%
Precision	0.1 to 0.7%
Range	up to 10 kg Pu (1.4 to 26 W)
Assay Time	4 h with end-point prediction

22.4 IN-PLANT CALORIMETERS

22.4.1 Mound Twin-Bridge Production Calorimeter

The twin-bridge production calorimeter (Figure 22.5) is designed for the assay of plutonium metal, oxide, and high-density scrap and waste (Ref. 11). The calorimeter is constructed for use in plant environments. Sample and reference cells are contained in a controlled constant temperature environmental water bath and are about 12 cm in diameter to accept standard cans in which plutonium materials are packaged. Calorimeters that can accommodate gallon-size cans of scrap and waste are also produced in this design. The instrument shown in Figure 22.5 has two twin-bridge calorimeters inside the water bath.

Each twin-bridge calorimeter may include a servo-controller for constant temperature operation, a sample pre-equilibrium chamber, or provisions for the use of end-point prediction techniques. These features can reduce assay time and increase throughput. All electrical measurements are made with bridge circuits or digital voltmeters and are fed directly into the computer data acquisition system. Standard resistors and voltage sources traceable to the National Bureau of Standards are used to ensure accurate measurements. Plutonium-238 heat standards are used for calibration.



Fig. 22.5 Mound twin-bridge production calorimeter. The water bath (left) contains two twin-bridge calorimeters. (Photograph courtesy of Mound Laboratory.)

At the Los Alamos Plutonium Processing Facility, two Model 102 twin-bridge calorimeters sharing a common water bath (Ref. 12) similar to that shown in Figure 22.5 are used to assay plutonium feed and product. To save space, sample pre-equilibrium chambers are not used. All assays are done for 8 h without end-point prediction. A measurement control program requires the weekly measurement of either a 400- or 870-g standard. Measurement control results for 1 yr show that the power determination has a precision of about 0.1% and a drift of about 0.1%. For large samples the effective specific power is determined with a precision of about 0.4 to 0.5% (1σ) by gamma-ray spectroscopy in count times of 1.5 to 2 h. The overall precision in plutonium mass determination is then about 0.5 to 0.7% (1σ) for large, homogeneous samples.* Performance data for the twin-bridge calorimeter are summarized in Table 22-5 (Refs. 11 and 12). Similar performance data has been reported for high-burnup plutonium with an effective specific power of about 14.5 mW/g (Ref. 13).

Table 22-5. Performance data for the Mound twin-bridge production calorimeter, assuming 2.3 mW/g and known isotopics (Refs. 11, 12, and private communication*)

Accuracy	0.1 to 0.2%
Precision	0.1 to 0.2%
Range	100 g to 2.5 kg (.23 to 5.8 W)
Assay Time	8 h

*Private communication from R. Blankenship and F. Hsue, Los Alamos National Laboratory.

By way of comparison, the Los Alamos facility also uses four over/under bridge calorimeters to provide increased throughput capability. These calorimeters are similar in appearance to the calorimeters described in Section 22.4.4 below. Two have 12.4-cm-diam sample chambers; the other two have 17.8-cm-diam sample chambers. The calorimeters are operated with end-point prediction techniques to reduce assay time. For samples containing 2 to 3 kg of plutonium, assay times are typically a little more than 3 h. Small or poorly packaged samples may require up to 8 h, and 100 g is the administrative lower limit on plutonium size to avoid longer assay times. Measurement control results for 1 yr show that these faster calorimeters have a measurement precision of about 0.2% compared to about 0.1% for the twin-bridge calorimeter.*

22.4.2 Mound Calorimeter for Simultaneous Isotopics Measurements

In-plant calorimetric assay requires both sample power measurement with a calorimeter and (for the computational method) determination of sample effective specific power from mass spectroscopy or gamma-ray isotopics measurements. For a series of samples, the most common approach uses sequential measurements of the plutonium isotopics

*Private communication from R. Blankenship and F. Hsue, Los Alamos National Laboratory.

by high-resolution gamma-ray spectroscopy followed by calorimetry. The isotopic measurement and the calorimetric measurement may also be performed simultaneously.

Figure 22.6 is an example of a simultaneous measurement system (Ref. 14). The calorimeter is a Mound transportable over/under bridge type (see Figure 21.5 of Chapter 21) with three layers of water (3.1 cm total), three layers of Plexiglas (2.1 cm total), 1.0 cm of epoxy, 0.6 cm of aluminum, and 0.1 cm of stainless steel between the sample and the gamma-ray detector. The transmission of 100- to 400-keV gamma rays through the calorimeter wall is about 20 to 40%. Because this calorimeter design uses Plexiglas, with a minimum of steel and aluminum, the transmission through the calorimeter is actually higher than the transmission through the x-ray filter on the face of the gamma-ray detector.

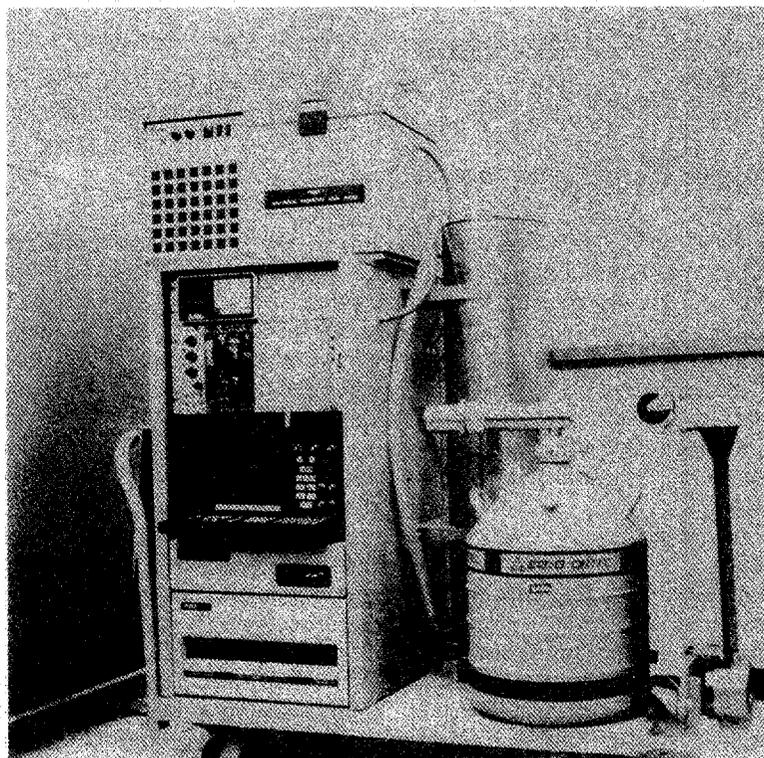


Fig. 22.6 Mound simultaneous calorimeter power measurement and gamma-ray isotopic composition measurement system. The side-looking germanium detector is placed against the calorimeter body during the measurement. (Photograph courtesy of Mound Laboratory.)

A typical assay time is 5 h, using end-point prediction techniques, with 0.3% uncertainty in the calorimeter power measurement (Ref. 14). Gamma-ray isotopic data were accumulated for the same time period as the power measurement. The overall assay uncertainty was 1% or less. The performance data are summarized in Table 22-6.

Both the calorimeter and the gamma-ray detector's multichannel analyzer are micro-processor controlled, and data analysis is coordinated by a minicomputer. This system is configured such that data from one sample can be analyzed while another sample is being measured. However, some loss of measurement precision is incurred in making simultaneous measurements because the sample cannot be placed directly against the gamma-ray detector. Sequential measurements are usually preferred to simultaneous measurements unless there are other overriding requirements.

Table 22-6. Performance data for the Mound simultaneous calorimetric assay system, assuming 2.3 mW/g (Ref. 14)

Overall accuracy	1.0%
Power determination accuracy	0.3%
Range	200 g to 4.4 kg (0.5 to 10 W)
Assay time	5 h with end-point prediction

22.4.3 Rocky Flats Production Calorimeters

Multiple-cell calorimeters have been developed at the Rocky Flats Plant for the assay of low-burnup plutonium oxide and scrap (Ref. 15). The calorimeters are of the twin-bridge type, with an air gap between the inner sample cell and the environmental water bath to help maintain a thermal resistance between them. Plutonium oxide cans containing approximately 1400 g are loaded into brass sample holders, which are placed into the measurement cells. Styrofoam plugs are used above and below the sample for insulation. The power output of each can is roughly 3 W. Shortly after the separation of americium from plutonium, 78% of the power output is from ^{239}Pu , 18% from ^{240}Pu , and the rest from ^{238}Pu and ^{241}Pu . The calorimetric assay of sample power is accurate to about 0.2%.

To handle the production load of samples requiring assay, three 8-unit calorimeters are available (Ref. 16). Each unit contains eight cells placed in a circle in a common water bath. Seven cells are available for sample assays, and one is used as a common reference cell.

For the new plutonium processing facility at Rocky Flats, a series of in-line calorimeters have been built by Mound Laboratory and installed under gloveboxes in the shipping and receiving area (Ref. 17). Twenty cells are connected in four banks of five cells each, with each bank having its own water bath. In each bank, one cell serves as the common reference cell for the other four sample cells. Each sample cell can equilibrate a 4-W sample in about 10 h. Overall measurement reproducibility is about 0.7% in all cells.

Calorimetric assay of plutonium scrap and waste encountered in the production process may be adversely affected by large container sizes, longer equilibrium times, or poorly known sample isotopic composition. An extreme example is found in the assay of impure salt residues that contain plutonium and up to 10% americium. In such residues

the americium may contribute 50 to 80% of the total sample power. A determination of plutonium content then requires a careful gamma-ray measurement of the americium to plutonium ratio in material that is very inhomogeneous and contains self-attenuating lumps of plutonium. This measurement has been performed by rotating and scanning the sample in a helical fashion during the isotopics measurement and by determining separate gamma-ray self-absorption correction factors for americium and plutonium (Ref. 18). The specific power is determined with a precision of about 1% and the sample power with a precision of about 0.2% (1σ). Although not yet established, the overall assay accuracy is better than that obtainable from passive gamma-ray or neutron assay, and has resulted in lower inventory difference biases (Ref. 18).

22.4.4 Mound Gradient Bridge Calorimeters

Mound Laboratory has provided four compact gradient bridge calorimeters for the plutonium scrap recovery facility at the Savannah River Plant (Ref. 19). The calorimeters are located in the sample assay room and are used to provide information on the mass of incoming plutonium metal and oxide shipments to the accountability computer. The four calorimeters (Figure 22.7) are connected by insulated water hoses to four separate temperature-controlled water reservoirs. Two of the calorimeters have outer dimensions of 31 cm diameter by 97 cm high, and accommodate standard recovery cans up to 12.4 cm in diameter. The other two calorimeters are 29 cm in diameter and 93 cm high, and only accommodate cans up to 11.0 cm in diameter, so that smaller samples can be assayed with shorter equilibration time.

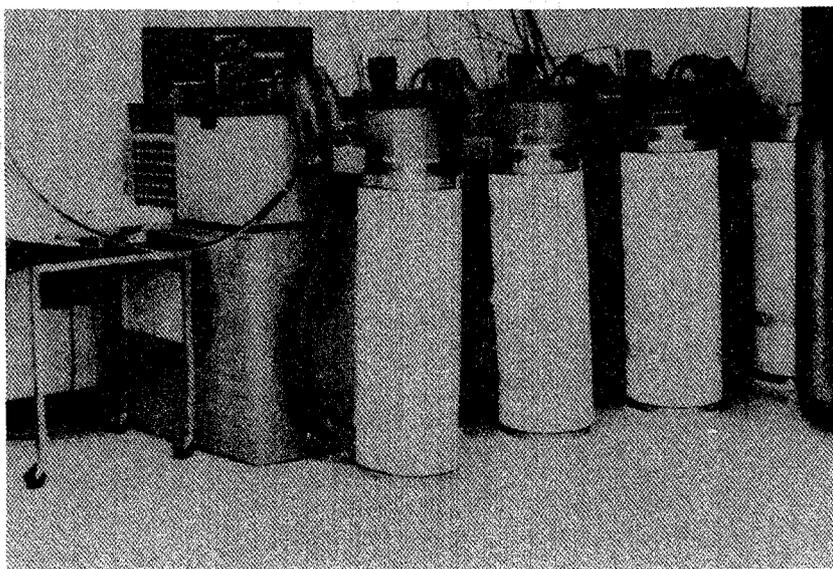


Fig. 22.7 Array of four Mound gradient bridge calorimeters, two with 11-cm-diam sample chambers and two with 12.4-cm-diam sample chambers.

The design of each calorimeter body (Ref. 20) is similar to the gradient bridge design shown in Figure 21.6 and described in Section 21.5.4 of Chapter 21. The thermal baffles above and below the sample chamber are made of Styrofoam. Sample weight is transmitted to the floor by a central post so that the accuracy of the bridge potential readings is not affected by strain in the windings. The sample is placed in a special stainless steel can with an O-ring seal that fits tightly into the sample chamber. If possible, air gaps between the sample and the inner wall of this "cal can" are filled with aluminum shot to reduce assay time, and air gaps above the sample are filled with crumpled aluminum foil to prevent air circulation. The time required to reach equilibrium depends on sample diameter, packaging, and the width of any air gap.

The four calorimeters are operated in the constant temperature servo-control mode (Ref. 20). The water bath temperatures are controlled to within 0.001°C, and the temperatures are checked every 2 min. The water temperature is selected such that the ratio of heating to cooling required from the controller is about 1.1. Each reservoir uses about one cup of distilled water per week. The electronics include five computer-controlled digital voltmeters, one for each bridge circuit output and one to monitor room temperature. Each heater winding is connected as a four-terminal resistor so that the voltage across the winding can be determined without errors resulting from power losses in the leads.

Operation of each calorimeter requires periodic baseline power runs to establish the empty chamber power level (Ref. 20). Baseline power runs typically require 2 to 2.5 h, less than actual sample assays, because there is no mass in the sample chamber. The baseline power level (near 15 W) is set at least 3 W greater than the estimated wattage for actual samples or calibration standards. The calorimeters have been provided with small $^{238}\text{PuO}_2$ heat source standards mounted in special cans. These standards are measured periodically to monitor instrument performance. Calorimeter performance can be affected if the calorimeter is bumped, if the lid is removed during an assay, or if fluctuations occur in room temperature and humidity, bath temperature, or power supply voltage. For plutonium scrap, assay time can vary widely depending on sample composition, packaging, size, and power. For a typical wattage range of 2 to 12 W, the accuracy of the calorimeter measurement is 0.2% (Ref. 21). Performance specifications for the gradient bridge calorimeters are summarized in Table 22-7.

Table 22-7. Performance specifications for the Mound gradient bridge calorimeters (Ref. 20 and 21)

Accuracy	0.5% at 2 W
Precision	0.1%
Range	500 g to 5 kg (2 to 12 W)
Assay time	4 h for product 8 to 16 h for scrap

22.5 FUEL ROD CALORIMETERS

22.5.1 Argonne One-Meter Fuel Rod Calorimeter

The Argonne one-meter fuel rod calorimeter is intended for the nondestructive assay of the plutonium content of unirradiated mixed-oxide fuel rods (Ref. 22). The prototype for this instrument is an earlier Argonne instrument used for the calorimetric assay of 6-in.-long fuel rods (Ref. 23). A four-meter fuel rod calorimeter has also been built (Ref. 24). Measurements with these instruments are relatively fast because of the small diameter of the sample chamber and the calorimeters are called "fast response" calorimeters.

This calorimetric assay system contains the following components (see Figure 22.8) (Refs. 25 and 26):

1. *Calorimeter box.* The one-meter fuel rod calorimeter is an isothermal gradient air calorimeter (See Section 21.5.4 of Chapter 21) that is operated in the servo-control mode (Section 21.6.3). It has three regions of controlled temperature: the 1-cm-diam sample chamber, the surrounding thermal shield and heating circuits, and the remaining volume of the box. These three regions are controlled to be at 33, 32, and 30°C, respectively.

2. *Preheater.* A preheater for heating rods prior to their injection into the measuring chamber is connected to the calorimeter measurement box. The preheater reduces assay time and provides additional space for holding rods larger than 1 m.

3. *Control console.* The control console contains the servo-control circuits, the temperature sensor controls, and other electrical measurement controls.

4. *Computer.* Data acquisition and analysis and hard-copy printout of results are controlled by a small portable computer.

The effective fuel rod length that can be accommodated by this instrument is 98 cm. Longer fuel rods must be assayed in 1-m segments. Along the axis, the response of the calorimeter to heat sources is nonlinear. A correction must be made for the length of the fuel column and its position (Ref. 22). Also, when the baseline power level is being determined, a dummy fuel rod must be inserted to simulate heat losses through the ends. Calorimeter operation can be checked by an internal electrical calibration or by calibration with electrically heated resistance rods.

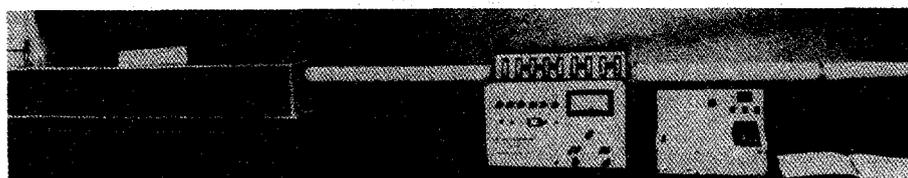


Fig. 22.8 Argonne Model IV fuel rod calorimetric system for one-meter fuel rods, showing the calorimeter (left), preheater (long white cylinder), and electronics packages. (Photograph courtesy of Argonne National Laboratory.)

Field test and evaluation exercises of the fuel rod calorimeter were conducted at several Belgian facilities in cooperation with the IAEA (Refs. 27 and 28). At one facility, four different geometries of fuel rods containing from 3.6 to 28.5 g of plutonium and having three different isotopic compositions were measured. All 12 fuel rods were preheated and then measured for 1 h, with each fuel rod measurement preceded by a 1-h baseline power measurement. The precision was 1 mW over the range of 10 to 135 mW. For rods with more than 50 mW of power, the observed accuracy was on the order of 3% (1σ) or less, which was within the errors of measurement and uncertainty in actual plutonium mass (Ref. 27).

Two more batches of 6 and 11 mixed-oxide fuel rods were measured at two other Belgian facilities. Some of the fuel rods were as long as 2.5 m; the inactive portion was kept within the preheater. For fuel rods of the type for which the instrument was designed, the calorimeter performed well, with a measurement precision of about 0.4%. For rods with low power output, the measurement data were inconclusive. Use of the one-meter fuel rod calorimeter for rods with power outputs less than 40 mW is not recommended because the power output would be less than 5% of the baseline power (Ref. 28). Performance data for the Argonne one-meter fuel rod calorimeter are summarized in Table 22-8.

Table 22-8. Performance data for the Argonne one-meter fuel rod calorimeter, assuming 5 mW/g and known isotopics (Refs. 27 and 28)

Accuracy	1 to 3%
Precision	0.4% or 0.2 g (1 mW)
Range	8 to 160 g Pu (40 to 800 mW)
Assay time	15 min to 2 h

22.5.2 General Electric Irradiated Fuel Assembly Calorimeter

The General Electric irradiated fuel storage facility near Morris, Illinois, has developed an in-basin calorimeter for underwater measurements of the heat generated by irradiated fuel assemblies (Ref. 29). The calorimeter is similar in size and shape to a water boil-off calorimeter developed earlier by Pacific Northwest Laboratory for above-water, hot cell measurements (Ref. 30). The General Electric calorimeter is operated in the unloading pit of the fuel storage basin at a depth of about 40 ft. Although the calorimeter was developed to provide heat generation information for planning future irradiated fuel storage needs, it was also possible to correlate the measured heat with fuel burnup.

The in-basin calorimeter (Figure 22.9) is 4.6 m long and consists of two concentric steel pipes (Ref. 29). The 41-cm-diam inner pipe forms the sample chamber, which can be fitted with inserts to support either boiling or pressurized water reactor fuel assemblies. The annular space between the two pipes contains 6 cm of urethane insulation to reduce heat transfer from the calorimeter to the basin water. Temperature measurements inside the sample chamber and outside the calorimeter are made with

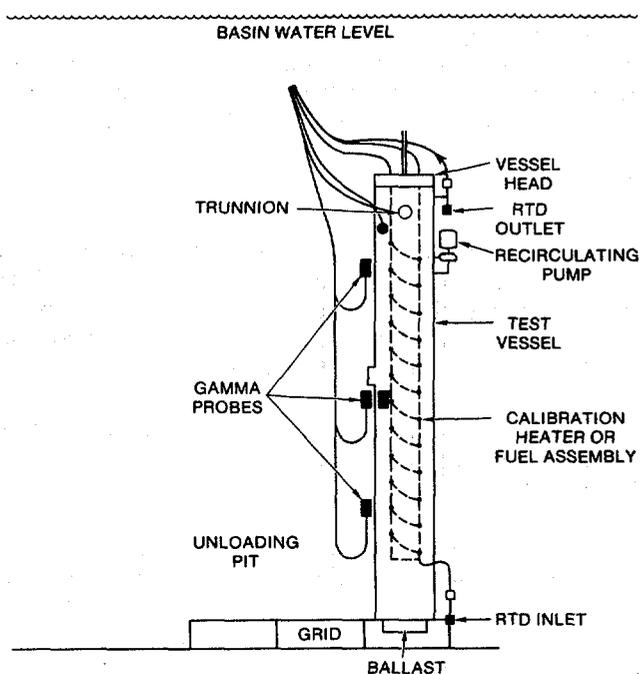


Fig. 22.9 General Electric-Morris Operations in-basin calorimeter for measurement of irradiated fuel assemblies in the unloading pit of the fuel storage basin—RTD = platinum resistance temperature detector. (Figure courtesy of General Electric-Morris Operations.)

platinum resistance temperature detectors. A recirculation pump maintains a homogeneous water temperature inside the sample chamber. Gamma radiation monitors are installed on the calorimeter to measure radiation heat losses and axial fuel assembly burnup profiles.

The calorimeter is usually operated with a constant temperature environment. After a fuel assembly is loaded into the calorimeter, a water-tight head is bolted on to seal the sample chamber (Ref. 29). The rise in internal water temperature is monitored over a 5-h period. Water temperature outside the calorimeter is usually stable to 0.1°F if water circulation is provided in the basin. The rate of change of internal water temperature (typically 2°F/h) is proportional to the thermal output of the fuel assembly. The calorimeter is calibrated with a 4-m-long pipe wrapped with electrical heater tape. All measurements are corrected for heater lead power losses, heat capacity variations between calibration and actual fuel, and gamma radiation heat losses.

Reference 29 reports a series of 24 measurements of 14 PWR fuel assemblies with operator-declared burnups of 26 to 40 GWd/tU and cooling times of 4 to 8 yr. The in-basin calorimeter measured thermal powers of 360 to 940 W with a precision of 1%. The

measured power was compared to that calculated from reactor fuel burnup codes. The agreement varied from 15% (if the codes assumed constant irradiation histories) to 1% (if the codes used the actual irradiation histories). For assemblies with the same cooling times, measured power was proportional to burnup to within about 3%.

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